THE JOURNAL OF GEOLOGY

VOLUME 60 MARCH 1952 NUMBER 2

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THE UNIVERSITY OF CHICAGO PRESS • CHICAGO

March 1952 Vol. 60, No. 2

THE JOURNAL OF GEOLOGY

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Claims for missing numbers should be made within the month following the regular month of publication. The publishers expect to supply missing numbers free only when losses have been sustained in transit, and when the reserve stock will permit.

Business correspondence should be addressed to The University of Chicago Press, Chicago 37, Ill.

Communications for the editors and manuscripts should be addressed to the Editor of THE JOURNAL OF GEOLOGY, Rosenwald Hall, The University of Chicago, Chicago 37, Ill.

The articles in this Journal are indexed in the *International Index to Periodicals*, New York, N.Y.

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Entered as second-class matter February 29, 1944, at the post office at Chicago, Ill., under the Act of March 3, 1879.
Acceptance for mailing at special rate of postage provided for in United States Postal Act of October 3, 1917, Section 1103, amended February 28, 1925, authorized July 15, 1918.

[PRINTED IN U.S.A.]

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"Mendocino Submarine Escarpment." By HENRY W. MENARD and ROBERT S. DIETZ

"Change of Melting Point of Diopside with Pressure." By HATTEN S. YODER, JR.

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——— and DUNSANY, A. J. (1929*a*) Early Ordovician faunas of South Dakota: U.S. Geol. Survey Bull. 444, pp. 1–76.

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VOLUME 60 NUMBER 2

THE JOURNAL OF GEOLOGY

March 1952

ORIGIN OF THE CONTRASTING MINERALOGY OF EXTRUSIVE AND PLUTONIC SALIC ROCKS[1]

O. F. TUTTLE

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ABSTRACT

Mineralogical differences between the quartz and feldspars of extrusive and plutonic acidic rocks are noted. The differences are believed to represent changes brought about in the minerals of plutonic rocks by inversion, unmixing, and recrystallization from an initial character similar to the quartz and feldspars of the corresponding extrusive rocks.

Equilibrium studies in the system orthoclase-albite-water provide quantitative information on the stability relations of the alkali feldspars. A single alkali feldspar is stable at high temperatures, but this phase becomes unstable at low temperatures and breaks up into a potash-rich and a soda-rich phase (perthite). Such an unmixing is common in the perthite-quartz granites. Granites of orthoclase-plagioclase-quartz (no perthite) may have the same chemical composition as the perthite-quartz granites, and it is suggested that the unmixing process may continue beyond the perthite stage to give nearly potash-free plagioclase and soda-free orthoclase. Such a process of unmixing and recrystallization may explain conflicting evidence for the order of crystallization in granitic rocks. It is believed that these processes—the result of changing stability relations at low temperatures—have produced significant changes in the mineralogy and textural relationships of the plutonic acidic rocks.

INTRODUCTION

Petrologists have long been aware of mineralogical differences between certain plutonic rocks and effusives having the same composition. For example, it has been noted that potash feldspar in rhyolites is usually sanidine, whereas the corresponding mineral in granites is microcline or orthoclase. It has been shown (Köhler, 1941) that the optics of the plagioclase feldspars are different in the plutonics and extrusives. Harker (1935) noted that the quartz of granites characteristically contains liquid inclusions, whereas the quartz phenocrysts of the corresponding rhyolites contain glass inclusions.

The significance of these differences does not appear to have been considered in any detail by petrographers. It is the purpose of this discussion to consider these and other mineralogical differences between certain plutonic rocks and their corresponding extrusives and to suggest certain implications of these differences.

Many trachytes low in lime contain one alkali feldspar as phenocrysts—sanidine or orthoclase—whereas the corresponding syenites may contain two feldspars—albite and orthoclase. Is this difference the result of different modes of origin for these two rock types, or is it caused by a lower temperature of crystallization in the case of syenites, as has been suggested by many petrographers? It is conceivable that the temperature of crystallization is lower in the syenites

[1] Manuscript received April 12, 1951.

during the final stages of crystallization, but surely the feldspar phenocrysts of a trachyte composed of 80 per cent anorthoclase phenocrysts crystallized at a temperature comparable to that of a syenitic feldspar which merely completed crystallization at depth. Another possibility, which will be explored here, is that the feldspars of plutonic rocks have changed from an initial character identical to the corresponding minerals of the extrusive equivalents by unmixing, inversion, and recrystallization to "plutonic-type" feldspars because of the changing stability relations at lower temperatures. If such a process can account for complete unmixing into separate phases, as would be required by the anorthoclase → albite + microcline relationship, then all intermediate stages (an "exsolution series") between anorthoclase and a mixture of albite and microcline, having the same total composition, should be demonstrable.

QUARTZ OF EXTRUSIVES AND INTRUSIVES

One of the most impressive features of the quartz phenocrysts of rhyolitic rocks is the presence of numerous large inclusions of glass. The inclusions are usually found in euhedral cavities, although somewhat rounded shapes are not rare. Each enclosure of glass normally contains a large spherical vapor bubble. These glass inclusions are common in the phenocrysts of extrusive rocks, but they are rare or nonexistent in the crystals of plutonic rocks. In the granites the quartz shows no evidence of these large glass inclusions. The glass would probably crystallize on cooling slowly in a plutonic environment, but the existence of the inclusion might be shown by large "negative crystals" composed of crystalline material, with some evidence of the former presence of a spherical vapor "bubble."

Petrofabric studies of quartz indicate that it is extremely susceptible to reorientation when subjected to deformation. For example, Turner (1948, pp. 192, 211) writes:

> . . . *quartz, the most sensitive mineral to deforming influence* [italics mine], was the only constituent of the rocks to readjust its orientation to the new movement picture. . . . This is consistent with a large body of general petrofabric evidence, suggesting that quartz responds more readily than micas in developing a state of preferred orientation during rock deformation.

Again, in discussing igneous versus metamorphic structures, Turner writes:

> It is usually difficult to differentiate between the igneous and metamorphic elements in the gneissic fabrics of such rocks, or even to distinguish between unmetamorphosed plutonic rocks whose gneissic structure is of purely igneous origin, and rocks that have been deformed in the solid state and hence possess a fabric which is, in part at least, metamorphic. In the absence of granulation, undulose extinction or marked preferred orientation in the crystals of quartz—*the most sensitive to deformation*—and parallel arrangement of mica flakes or prisms of hornblende may safely be interpreted as due to magmatic flow. Such cases are relatively rare, however, since the forces which bring about sustained flow of a highly viscous, largely crystalline magma are usually sufficiently powerful to cause rupture and incipient granulation of some of the grains of quartz and feldspar in the last stages of magmatic flow. *More often than not, the deforming forces continue to operate after crystallization is complete, and consequently then imprint a truly metamorphic fabric upon the quartz* and perhaps upon other minerals as well [italics mine].

It is the writer's experience that quartz of granites commonly, if not always, shows undulose extinction, and evidence of granulation is often present. Preferred orientation of quartz is the rule in granitic rocks, although such an equant mineral would not be expected to be oriented by flow. The quartz of granites commonly has the same orientation pattern as the

quartz in surrounding metamorphic rocks—a relation which appears to demand postcrystallization reorientation. The quartzes of many granites, particularly the coarse-grained types, actually look like "fragments" of quartzite.

Studies on the inversion of quartz between temperature of formation and the inversion temperature. If this relation holds for the granites and rhyolites, then the quartz of granites must have crystallized or recrystallized at some lower temperature to give the relation shown in figure 1. It is reasonable to as-

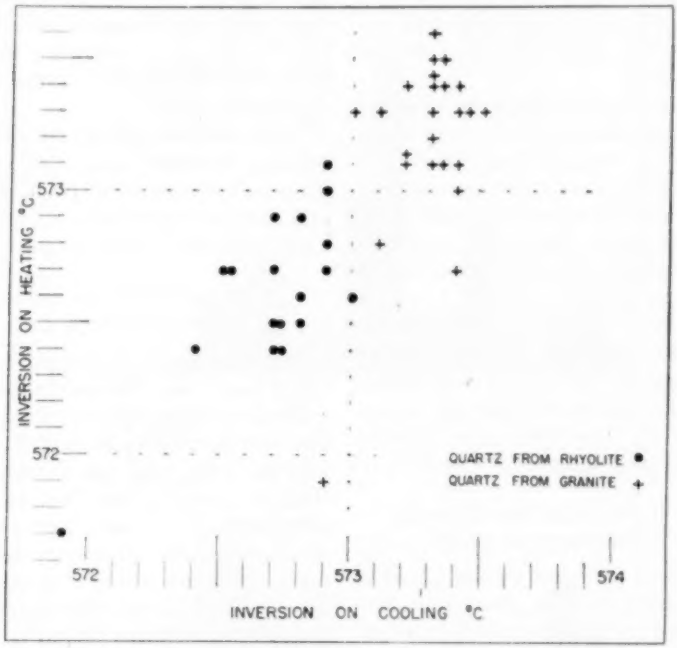


FIG. 1.—Graph showing the relation between beginning of the high-low inversion on heating and on cooling in quartz from 23 granites and 18 rhyolites (after Keith and Tuttle, 1950).

(Tuttle, 1949*b*; Keith and Tuttle, 1950) show that, in general, quartz from granitic rocks goes through the high-low inversion at a slightly higher temperature than do quartz phenocrysts of rhyolites. This difference is illustrated in figure 1, which is a plot of the temperatures of beginning of transformation of quartz on heating through the inversion against the beginning of transformation on cooling through the inversion. The work also suggests that there is an inverse relation

sume that the quartz of granites completed crystallization at a somewhat lower temperature than "rhyolitic" quartz and that therefore part of it may have grown at a temperature below that represented by the phenocrysts of extrusive rocks. In such a case a portion of the granite-quartz should show inversion characteristics similar to those of rhyolite-quartz. No evidence of "rhyolitic" quartz was discovered, despite the fact that the method used in the quartz

studies can detect less than 5 per cent of one quartz mixed with another of contrasting inversion characteristics!

Quartz of granitic rocks often contains numerous minute rutile needles that occur predominantly along three crystallographic directions. This suggests that TiO_2 has unmixed from the quartz, a reaction which might be expected during recrystallization at low temperatures.

All in all, there is convincing evidence that the quartz of extrusive rocks is not the same as quartz from "plutonic" rocks. The fact that quartz is reluctant to admit foreign atoms into solid solution has made it difficult to establish chemical differences which would be invaluable in deciding whether or not recrystallization had taken place. However, evidence from the inclusions, the inversion, and the petrofabric data strongly suggests recrystallization as an explanation for the differences between "plutonic" and "extrusive" quartz.

PLAGIOCLASE FELDSPARS IN EXTRUSIVE AND PLUTONIC ROCKS

Plagioclase feldspars are not found as major constituents of phonolites, trachytes, and rhyolites unless the lime content is relatively high. If present as phenocrysts in these rocks, the plagioclase is characteristically zoned. In contrast, plagioclases in the corresponding plutonic rocks usually lack zoning, and, if it is present, the compositional variations are not so pronounced as in the plagioclase of extrusive rocks.

A most valuable contribution to our knowledge concerning the plagioclase feldspars has been the recognition by A. Köhler (1941) that the optical properties of "plutonic" plagioclase feldspars differ significantly from those of the feldspars of extrusive rocks. Köhler and co-workers have published new sets of determinative curves and tables for the plagioclases, one set for the high-temperature group and another for the low-temperature "plutonic" series. In the case of the soda-rich plagioclase feldspars, it has been established (Tuttle and Bowen, 1950) that two distinct modifications exist; high-temperature albite has been found only in extrusive rocks and meteorites, whereas the low form is found in plutonic rocks.

Again, as with quartz, the differences between "plutonic" plagioclase and high-temperature "extrusive" plagioclase indicate that inversion and recrystallization have taken place in the plutonic rocks.

ALKALI FELDSPARS OF EXTRUSIVE AND PLUTONIC ROCKS

The quartz and plagioclase feldspars, which have been discussed above, show evidence of being different in the extrusive and plutonic rocks, and it could be inferred from these relationships alone that the plutonics have had an entirely different mode of origin from the extrusive equivalents. However, the alkali feldspars, which will now be considered, show a gradational relationship which links the end-stages shown by quartz and plagioclase and strongly supports the thesis that extrusives and plutonics have a common ancestry. The lack of evidence for gradation in the quartz and plagioclase is due to our inability to detect intermediate stages as such.

MINERALOGY

A striking difference between the potash-bearing feldspars of salic extrusive rocks and the corresponding plutonics is evidenced by the numerous descriptions of these rocks throughout the petrographic literature. Sanidine, anorthoclase, soda-orthoclase, or soda-

microcline are the usual designations for the feldspar phenocrysts of the extrusives, whereas the potash-bearing feldspar in the plutonics is invariably identified as microcline or orthoclase. These

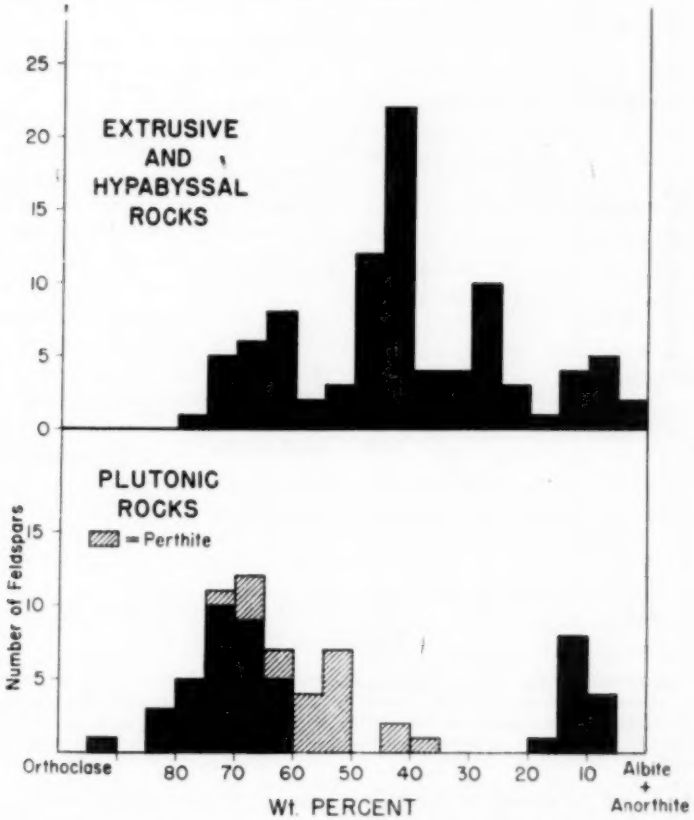


FIG. 2.—Histograms showing the relation between number of analyses and composition of alkali feldspars in plutonic and extrusive + hypabyssal rocks.

descriptions suggest that the feldspar phenocrysts of the extrusives are richer in soda than the plutonic microclines or orthoclases. In figure 2 the compositions of extrusive and hypabyssal feldspars are contrasted with those from the corresponding plutonic rocks. Extrusive feldspars cover the range from Or_{75} to Or_0, with a great majority falling between Or_{40} and Or_{50}. This striking concentration is somewhat exaggerated by rather a large number of analyses of feldspar from certain Norwegian and Mediterranean rocks which have been extensively studied by many investigators. However, even without these analyses the histogram would show essentially the same relation, that is, feldspars ranging from Or_{72} to Or_0 with no gap in the series. On the other hand, the plutonic feldspars show a gap in the

range of compositions and form two groups with the maximum for the potash-rich feldspars near Or_{72} and that for soda feldspar near Or_{12}. (This agrees with an earlier compilation by Tschirwinsky [1931], who found the average of 82 orthoclases from granites to be Or_{72} Ab_{25} An_3.) This figure of Or_{72} is certainly too low for the orthoclase content of the potash feldspar in granites, as many of the analyses were undoubtedly made on fine perthitic material. The range Or_{40}–Or_{80}, which represents nearly 64 per cent of the analyses of extrusive feldspars, is represented by less than 20 per cent of the analyses of plutonic feldspars, and each analysis in this 20 per cent represents a perthitic intergrowth of two feldspars.

These relations suggest that many of the potash-bearing feldspars present in the salic plutonic rocks have changed from an initially soda-rich composition by unmixing. Equilibrium studies in the system albite-orthoclase-water (Bowen and Tuttle, 1950) provide quantitative information on the subsolidus relations of these alkali feldspars. They show that a single feldspar is stable at the high temperatures of trachytic magmas but that it becomes unstable at lower temperatures and breaks up into two phases, albite and orthoclase, when heated for a prolonged period below the solvus (fig. 3, *ABC*). Such prolonged heating is expected in the slow-cooling plutonic rocks but not in the extrusives and small hyp-

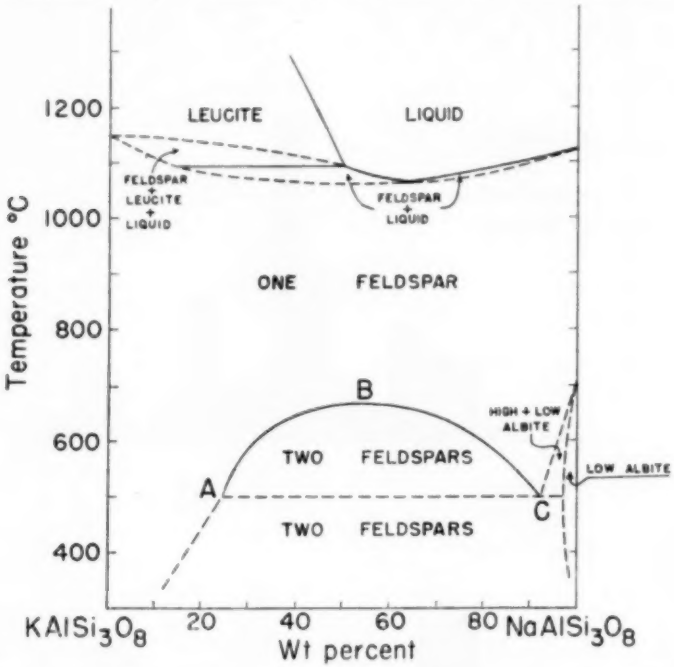


FIG. 3.—Equilibrium diagram for the system $KAlSi_3O_8$-$NaAlSi_3O_8$ (Bowen and Tuttle, 1950), showing the probable relations of high and low albite. The solvus (*ABC*) was located by crystallizing mixtures in the two-feldspar region and determining the composition of the crystals by means of the X-ray. Mixtures crystallized as a single homogeneous phase at temperatures above the solvus unmixed on heating in the two-feldspar field.

abyssal intrusives. The amount of soda feldspar that can be unmixed from the potash-bearing phase depends on the initial composition, and this, in turn, is dependent largely on the soda-potash-lime relationship in the parent-rock.

SIGNIFICANCE OF THE MINERALOGICAL DIFFERENCES BETWEEN SALIC EXTRUSIVES AND INTRUSIVES

Petrographers have long noted perthitic intergrowths of the alkali feldspars in the plutonic salic rocks and have deduced that they originate by unmixing from a pre-existing homogeneous feldspar. To be sure, some investigators have made other suggestions, such as eutectic intergrowth and metasomatism, but, in general, opinion seems to lean heavily toward exsolution as the most likely process. Heating experiments, such as the excellent study carried out by Spencer (1938, *passim*), leave little doubt as to the unmixing origin for at least some perthites. Recently it has been possible to determine experimentally the phase relationships in perthite by using water vapor as a flux (Bowen and Tuttle, 1950). These relationships are summarized in the equilibrium diagram reproduced in figure 3. It was found that at temperatures above the solvus (fig. 3, *ABC*) mixtures of potash and soda feldspars crystallize to one homogeneous feldspar, whereas at lower temperatures two feldspars form side by side. The composition of the two feldspars varies with temperatures below 660° C., as shown by the solvus. The composition of the potash-rich phase changes along *BA* and that of the soda-rich phase along *BC* of the solvus. Also, feldspars formed above 660° will, on prolonged heating below 660°, *break up into two feldspars.*

The possibility that unmixing and recrystallization may continue beyond the perthite stage, resulting in elimination of the potash feldspar from the soda feldspar, must also be considered. If such an extension of the unmixing series can be established, disturbing features in the granites, such as conflicting evidence for the order of crystallization as deduced from grain relationships, might be resolved. If, however, it can be demonstrated that salic plutonic rocks containing relatively pure potash feldspar and soda feldspar have not formed by unmixing, then a useful criterion for an origin other than magmatic is indicated, since equilibrium relations between the potash and soda feldspars with water leave little room for the possibility of such pure feldspars crystallizing together from trachytic magma. (A single homogeneous feldspar was obtained even with upward of 10 per cent water and 3,000 kg/cm^2 pressure.)

The possibility that small amounts of lime drastically alter the equilibrium relations must, of course, be considered, as plagioclase phenocrysts are formed in lime-bearing rhyolites and trachytes. The study by Larsen (1938) on San Juan lavas indicates, however, that the potash feldspar is rich in soda even when plagioclase is present. The probable influence of lime will be considered later.

THE EXSOLUTION SERIES

The alkali feldspars found in salic rocks can be grouped into an "exsolution series" consisting of seven types. Of these seven types, five can be recognized optically. The seven types are: (1) sanidine or anorthoclase (homogeneous crystals), (2) sub-X-ray perthite, (3) X-ray perthite, (4) cryptoperthite, (5) microperthite, (6) perthite, and (7) albite and orthoclase (or microcline). From consideration of published descriptions and microphotographs and studies of numerous thin-sections, the thicknesses of lamellae in the different types are as

follows: (2) probably less than 15 Å in $\perp(\bar{2}01)$ direction, (3) $< 1\,\mu$, (4) 1–5 μ, (5) 5–100 μ, (6) 100–1,000 μ (1 mm.).

Studies of sanidines and anorthoclases indicate that perthitic intergrowth exists in one of the phases which is so minute that the X-ray spectrometer fails to detect its presence (201 spacing absent). Evidence for the existence of such a perthite is obtained by comparing the composition as determined by the X-ray (Bowen and Tuttle, 1950, p. 493) with that indicated by chemical analysis. Anorthoclase from Victoria, Australia, for example, has a composition by chemical analysis of $Ab_{77}Or_{23}$, whereas the X-ray method gives $Ab_{93}Or_7$ with no evidence of orthoclase being present as an unmixed phase. On heating at a temperature above the solvus and redetermining the composition by X-ray, a value of $Ab_{76}Or_{24}$ is obtained, indicating that orthoclase is present before heating in sub-X-ray units and that the heat treatment dissolved the unmixed orthoclase.

Many crystals described as sanidine which appear homogeneous optically under the highest magnification are shown by X-ray studies to consist of two phases, one nearly pure albite and the other nearly pure orthoclase. These are classified above as "X-ray perthites," type 3. Types 4, 5, and 6 have long been recognized and described by petrographers.

This series represents a decreasing temperature plus increasing time scale and therefore should prove of value in studying the thermal history of certain igneous rocks. The position in the exsolution series is related to the rate at which the feldspar cooled and this, in turn, if there is no extraneous source of heat, to the size of the intrusions. For example, X-ray perthites, type 3, would be expected only in explosive volcanics and associated tuffs, whereas cryptoperthites would be expected in small hypabyssal dikes and sills and chilled borders of larger intrusives. In the absence of reheating, microperthite and perthite should appear only in small plutons, whereas large plutons of granites and syenites should, in general, show orthoclase and plagioclase, not perthitically intergrown, the orthoclase having a plagioclase content less than Ab + An = 20. These relations will hold only if the intrusion has a soda-to-potash ratio which permits formation of sanidine or anorthoclase of the composition appropriate for unmixing at the lower temperatures. Under favorable conditions it should be possible to find chilled borders showing all gradations from anorthoclase at the contact to cryptoperthite, microperthite, perthite, and plagioclase + orthoclase, appearing progressively in passing from the contact toward the center of the intrusive. Actually, gradations from anorthoclase to perthite are not uncommon, and gradations from cryptoperthite to perthite are common. Phemister (1926), for example, *described the complete exsolution series* in the perthosites of Scotland. He wrote (p. 25): "A study of the feldspars of these rocks indicates that the perthitic intergrowth is not a stable condition, for every variation from cryptoperthite to independent crystals of soda and potash-feldspars is met." Also, "Segregation of perthite into its components is usual in

PLATE 1

Granite from Modoc, Ontario, which is essentially perthite and quartz. No plagioclase is present outside the perthite grains.

Granite from Modoc, Ontario

Perthite-quartz granite from Quincy, Massachusetts

rocks which have undergone recrystallization either as an effect of deformation or because of the presence of hot solutions." The exsolution series is evidently represented in these perthite-bearing rocks, but its relation to the more quickly cooled portions of the rocks is not discussed.

Granite from Quincy, Massachusetts, appears to show some evidence of an intermediate step in the perthite→orthoclase + plagioclase exsolution stage. Plate 2 is a photograph of a stained section of the Quincy granite, showing perthite and quartz, with each perthite grain sheathed in fine-grained plagioclase. Such interstitial plagioclase may owe its origin to exsolution. Unmixing, in which the exsolved phase segregates at grain boundaries, has been recognized and described often in alloys and occasionally in sulfides. In contrast, the Modoc, Ontario, granite, shown in plate 1, consists essentially of perthite and quartz, with little or no interstitial plagioclase.

PETROGRAPHIC EVIDENCE FOR THE EXSOLUTION SERIES

The transition from perthite to orthoclase plus albite[2] stage is difficult to establish, once the perthite stage is passed, as the accepted evidence for unmixing is lost. However, a grain relationship, which has apparently escaped previous notice, supports the hypothesis. This is the relation of "secondary" albite to the boundary between potash feldspar and plagioclase feldspar in certain granites. If the potash feldspar of the granites is the source of this so-called "late-stage" or "deuteric" albite, then the albite should show a definite preference for the boundary between potash and soda feldspar (pls. 3, 4).

At the suggestion of F. Chayes of this laboratory, a simple test was set up to test the strength of this association. Two thin-sections of each granite to be investigated were traversed at intervals such that the same grain would not be encountered more than once. As each of six possible boundaries was passed (Or-Or, Pl-Pl, Qtz-Qtz, Or-Pl, Or-Qtz, Pl-Qtz), a record of the type of boundary and the presence of secondary albite was recorded for 100 grain contacts. Contacts involving other constituents were ignored. Results for six granites are shown in table 1. *Nearly all the secondary albite lies at the orthoclase-plagioclase boundary.* At least a portion of the plagioclase of these granites appears to be derived from the potash feldspar by unmixing, but it is not clear how much of the plagioclase has formed in this fashion.

The existence of the exsolution series may be inferred from the contrasting modes of rocks having essentially the same chemical composition (heteromorphic). Table 2 is such a series. The series from sanidine to perthite is well represented. The final stage from perthite to albite + microcline is more difficult to establish. The fact that rocks having identical composition may consist largely of perthite in one case and a mixture of albite and microcline in another

[2] The plagioclase component of perthite is commonly oligoclase and in some cases may be as calcic as andesine.

PLATE 2

Perthite-quartz granite from Quincy, Massachusetts. *Q* = quartz; *O* = fine-grained plagioclase feldspar sheathing the perthite grains. This plate is believed to represent an intermediate stage between a perthite-quartz granite showing no interstitial plagioclase (pl. 1) and a granite consisting of microcline, plagioclase, and quartz as discrete grains with little or no perthite material (pls. 3, 4). The interstitial plagioclase is believed to have unmixed from the potash-bearing phase. Potash feldspar stained.

is not in itself convincing evidence for exsolution. However, when the equilibrium studies of the system orthoclase-albite-water, the rate at which exsolution may take place in the laboratory, the microscopic evidence from the relation of "secondary" albite to the orthoclase-plagioclase boundary, and the various other textural relations found in granites are considered, the probability of complete unmixing as a factor in the evolution of some granites must be considered. The probability of microcline and albite, of the composition found in many granites, crystallizing from a granite magma side by side is exceedingly remote. The association of relatively pure albite and microcline suggests temperatures so low that an origin other than magmatic is indicated unless exsolution can be shown to be operative. Perthite, when composed of nearly equal amounts of albite and microcline, is evidence per se of high temperature and suggests a magmatic history for the rocks concerned.

Some heteromorphic pairs of granites are shown in table 3. The first pair (Westerly and Pikes Peak granites) are so similar in composition that there is little doubt that the perthite in the Pikes Peak is represented by microcline and plagioclase in the Westerly granite.

TABLE 1

RELATION BETWEEN ALBITE RIMS ON PLAGIOCLASE AND ORTHOCLASE-PLAGIOCLASE BOUNDARY IN SOME GRANITIC ROCKS

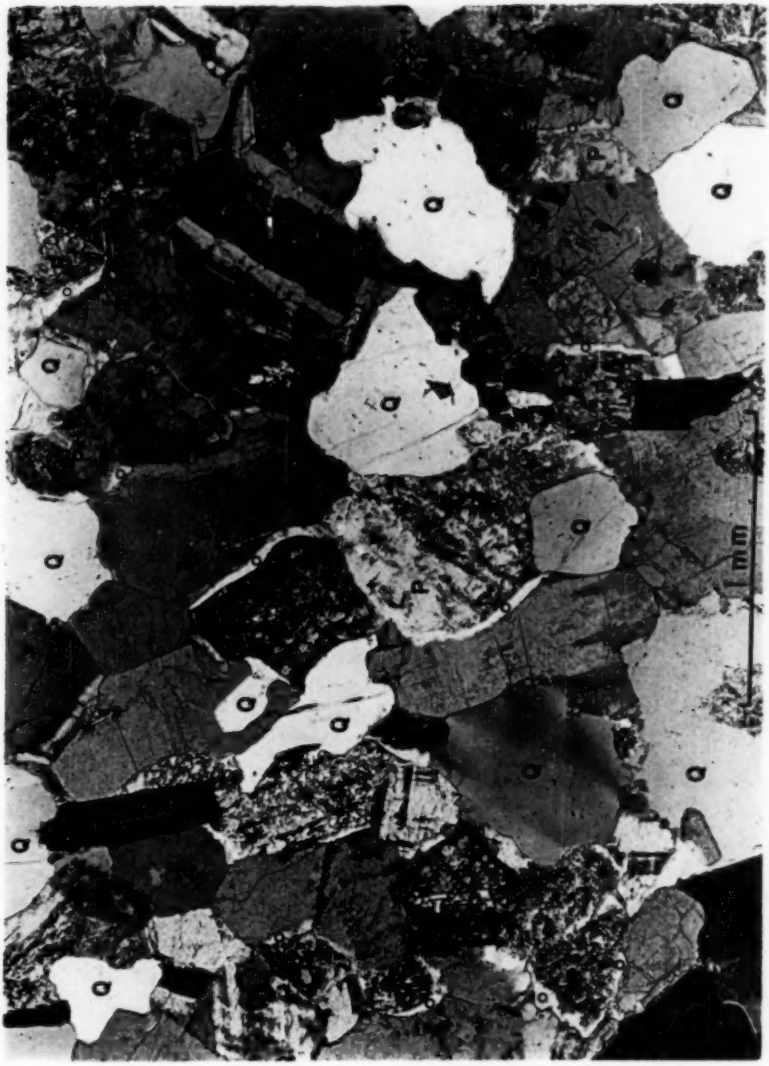
Locality	No. of Contacts	Or-Pl	Or-Or	Or-Q	Pl-Pl	Pl-Q	Q-Q	Total
Pownal, Maine	Total	63	14	83	4	31	5	200
	With secondary ab.	50	0	0	0	0	0	
Bradford, Rhode Island	Total	61	10	42	25	51	13	200
	With secondary ab.	51	0	0	4	1	0	
Barre, Vermont	Total	56	7	31	38	51	17	200
	With secondary ab.	48	1	0	1	0	0	
Westerly, Rhode Island	Total	66	13	53	9	51	9	200
	With secondary ab.	56	2	0	0	0	0	
Llano, Texas	Total	54	25	72	7	26	16	200
	With secondary ab.	48	0	0	0	0	0	
Woodbury, Vermont	Total	26	4	11	18	26	16	100
	With secondary ab.	20	0	0	1	1	0	

HOW MUCH OF THE PLAGIOCLASE IN GRANITES UNMIXED FROM THE POTASH FELDSPAR?

Rhyolites contain phenocrysts of plagioclase in addition to sanidine when the lime content is appreciable. We must

PLATE 3

Westerly, Rhode Island, granite illustrating the relation of sodic plagioclase to the boundary between potash feldspar and plagioclase feldspar. Q = quartz; P = plagioclase; O = sodic plagioclase rim between orthoclase and plagioclase only. Note the absence of the rim at quartz-plagioclase, orthoclase-quartz, and plagioclase-plagioclase boundaries. This relation is believed to indicate that the plagioclase rim unmixed from the potash feldspar.



Granite from Westerly, Rhode Island

Granite from Pownal, Maine

conclude, then, that not all the plagioclase of granites owes its origin to unmixing. On the other hand, many granites contain no plagioclase at all except as a component of perthite. Figure 4 has been compiled to show these relationships. The CaO, Na_2O, and K_2O proportions in the granitic rocks (including granodiorites) are shown, together with a symbol for the amount of plagioclase in the rock. The dotted curve *AB* divides the chart into two fields. Granites whose compositions place them above the curve contain both plagioclase and orthoclase. The area under the curve is particularly interesting, as granites falling in this region may contain *as much as 35 per cent plagioclase or none at all*. It is concluded that, in granites represented by points under the curve, all the plagioclase may have unmixed from a soda-rich (and, in some cases, rather lime-rich) anorthoclase, unless granites containing plagioclase have an entirely different mode or origin from that of perthite-quartz granites.

The effect of lime on the composition of the potash feldspar phenocrysts is well illustrated by the excellent studies of Larsen (1938) on the San Juan ex-

TABLE 2

HETEROMORPHIC SERIES—TRACHYTES

	Aegirite-Trachyte*	Soda-Trachyte†	Nordmarkite‡	Solvsbergite§
SiO_2	60.74	63.43	63.32	64.92
TiO_2	1.05	1.19	0.52	.....
Al_2O_3	15.58	16.31	18.75	16.30
Fe_2O_3	3.66	2.04	1.24	3.62
FeO	3.55	3.14	1.77	0.84
MnO	0.26	0.04	0.11	0.40
MgO	0.38	0.78	0.71	0.22
CaO	0.97	1.70	2.03	1.20
Na_2O	6.59	6.71	6.04	6.62
K_2O	4.62	4.31	4.88	4.98
H_2O^-	1.21	0.26	0.06	0.50
H_2O^+	1.20	0.18	0.36	
P_2O_5	0.07	.....	.....	.....
	Modes			
Sanidine	83	.....	.....	.....
Cryptoperthite	.....	83	.....	.....
Microperthite	.....	.....	83	.....
Microcline	.....	.....	.....	29
Albite	.....	.....	.....	50
Others	17	17	17	21

* Lake Naivasha, Kenya, East Africa. M. H. Hey, analyst (see Smith, 1931, p. 227).
† Montagna Grande, Pantelleria, Italy. H. S. Washington, analyst (1913, p. 688).
‡ Pleasant Mountain, Maine. Kameda, analyst (see Jenks, 1934, p. 330).
§ Solvsberget, Gran Parish, Norway. L. Schmelck, analyst (see Brögger, 1894, p. 78).

PLATE 4

Pownal, Maine, granite. *Q* = quartz; *P* = plagioclase; *O* = sodic plagioclase rim between microcline and plagioclase. The rim is absent at plagioclase-quartz, microcline-quartz, and plagioclase-plagioclase boundaries, an indication, it is believed, that the rim owes its origin to unmixing from the potash feldspar.

trusives. Figure 30 of Larsen's paper (p. 422) shows that in rocks poor in lime the sanidine contains more albite than orthoclase and that the composition of the crystals in such cases is essentially the same as that of the normative feldspar; hence it is expected that these rocks would crystallize only one feldspar. On the other hand, when the normative anorthite is 10 per cent or more, two feldspars are present, the potash phase containing approximately 35–40 per cent albite and the plagioclase containing 25–40 per cent anorthite. Crystallization of plagioclase containing 25–40 per cent anorthite will rapidly remove lime from the liquid, and, unless perfect equilibrium is attained, the composition of the liquid will soon become such that only one feldspar (sanidine rich in albite) will be precipitated. The ubiquitous zoning in the plagioclase phenocrysts of lavas indicates that equilibrium is rarely attained, and therefore sanidine rims around plagioclase, as well as sanidine phenocrysts with potash-rich cores and soda-rich mantels, should be common. The hypabyssal dike rocks show just such relations. For example, Warren (1913) described phenocrysts with mantels of perthite in the granite porphyries of Quincy, Massachusetts. Ofte-

TABLE 3

HETEROMORPHIC GRANITES: THREE PAIRS OF GRANITES HAVING SIMILAR CHEMICAL COMPOSITIONS WITH GREATLY DIFFERENT MODAL COMPOSITIONS

	Westerly, Rhode Island*	Pikes Peak, Colorado†	Palabora, Eastern Transvaal‡	Quincy, Massachusetts§	Hudiksvall, Sweden‖	Adirondacks, New York#
SiO_2	72.34	73.51	73.65	71.41	71.73	75.98
TiO_2	0.26	0.18	0.17	0.38	0.32	0.26
Al_2O_3	14.34	13.28	14.40	12.74	14.85	11.92
Fe_2O_3	0.68	0.94	0.31	1.75	0.23	1.24
FeO	1.13	0.97	0.51	2.33	1.72	1.26
MnO	0.02	trace	0.004	0.10	0.04	0.04
MgO	0.37	0.05	0.10	0.06	0.73	0.29
CaO	1.52	1.11	0.97	0.85	1.27	0.18
Na_2O	3.37	3.79	4.46	4.59	3.45	3.67
K_2O	5.47	5.22	5.00	5.00	5.06	4.69
H_2O^-	0.06	0.16	0.27	0.10	0.12	0.04
H_2O^+	0.30	0.62	0.19	0.56	0.43	0.14
	Modes					
Quartz	27.5	27.6	27	23	31	38
Microcline	35.4	.....	35	.....	23	.....
Plagioclase	31.4	.....	35	.....	36	6
Perthite	.....	68.3	.....	66	.....	52
Others	4.5	3.4	3	11	10	4

* J. H. Scoon, analyst (unpublished). The writer is indebted to C. E. Tilley for this analysis. F. Chayes, mode (1952).

† W. F. Hillebrand, analyst. O. F. Tuttle, mode (see Matthews, 1900, locality 2530). (Also BaO = trace; Li_2O = trace; F = 0.55.)

‡ S. J. Shand, analyst (1931, p. 86). (Also P_2O_5 = 0.02; BaO = 0.06.)

§ C. H. Warren, analyst (1913, p. 235). (Also P_2O_5 = 0.22; CO_2 = 0.40; ZrO_2 = 0.10.)

‖ Almström, analyst (see Von Eckermann, 1928).

A. Willman, analyst (see Buddington, 1939, p. 148). (Also CO_2 = 0.06; P_2O_5 = 0.02; BaO = 0.03.)

dahl (1948) also described sanidine phenocrysts with perthite rims and plagioclase phenocrysts with mantels of sanidine. Buddington (1939, p. 85 and *passim*) has described Adirondack syenitic and granitic rocks containing perthite with cores of plagioclase. In these rocks it is apparently common to find nearly all the plagioclase either as lamellae of perthite or as cores of perthite crystals.

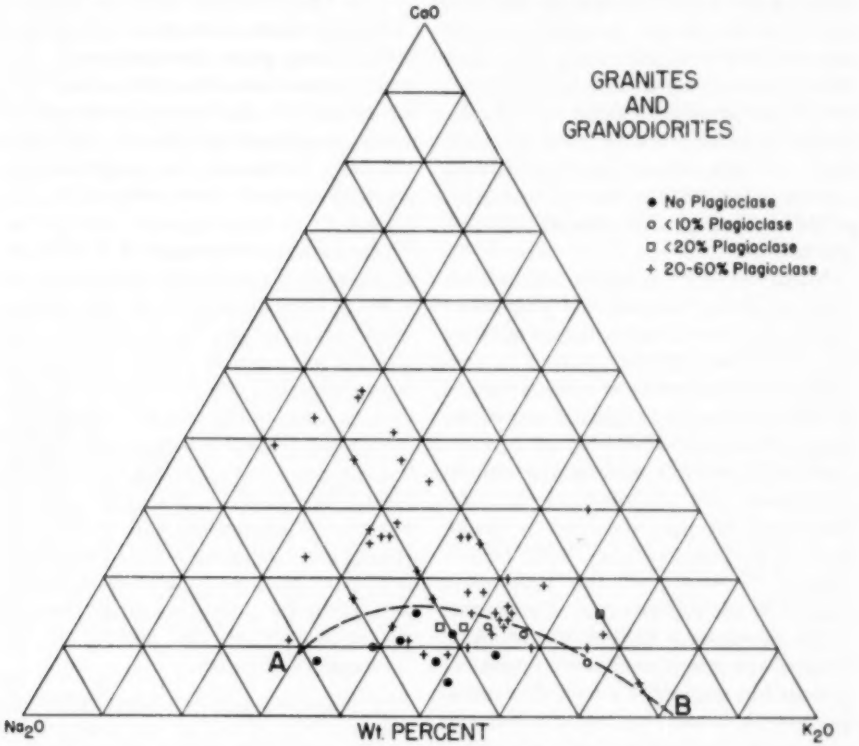


FIG. 4.—Relation between CaO-Na_2O-K_2O content and modal plagioclase of 57 granites and granodiorites. All values shown are taken from Johannsen (1932), vol. 2. Those rocks high in iron, magnesia, and lime are omitted.

Chayes (1950) has examined in great detail a suite of granites in which plagioclase is a major constituent and perthite rare, and he concludes that a magmatic history is demanded to explain the relations found. Some of these granites have a chemical composition which places them very close to or below the line *AB* of figure 4. Have these granites a different history from those of the same composition consisting of perthite and quartz? It is conceivable that extreme fluxing by volatiles would lower the liquidus sufficiently to permit simultaneous crystallization of orthoclase and plagioclase, in which case there would be two types of magmatic granites: (1) "wet" granites, consisting of Or-Pl-Qtz in nearly equal amounts, and (2) "dry"

granites, consisting of perthite and quartz. Goranson's (1932) studies on the melting of granite suggest that the amount of water necessary to lower the liquidus to a temperature below 660° (highest temperature of the solvus, fig. 3) is over 6 per cent, if we assume that the solvus is not changed greatly by 1 per cent of CaO. It might be expected that this amount of water, if originally present in the granites studied by Chayes, would manifest itself in some manner, such as pronounced contact effects, numerous pegmatites, etc. In contrast, perthite-quartz granites should lack such effects.

Many of the granites studied by Chayes show "secondary" plagioclase (see above) at the orthoclase-plagioclase boundary (see table 1), and it is concluded from this that at least a portion of the plagioclase unmixed from the potash feldspar. There is no obvious method of determining how much of the plagioclase not represented by this plagioclase rim owes its origin to exsolution. If a chilled contact could be discovered, a study of the progressive change in the feldspar near the contact might throw some light on the problem. Some of the granites whose compositions lie near but above the dotted line *AB* of figure 4 have been examined and found to show "albite" rims between the orthoclase and plagioclase, indicating that for these rather high-lime rocks at least a portion of the plagioclase owes its origin to unmixing from the potash feldspar.

It should be noted that the Al_2O_3 in the analyses shown in table 3 is greater in the plagioclase-microcline-quartz granites than in the perthite-quartz granites. This consistent difference may be significant. Alumina present in these low-lime acidic rocks in excess of the Al_2O_3 to SiO_2 ratio in soda and potash feldspars may lower the liquidus appreciably and thus be effective in permitting two feldspars to crystallize at the liquidus.

GRAIN RELATIONSHIPS IN GRANITIC ROCKS AND THE ORDER OF CRYSTALLIZATION

Interpretation of an order of crystallization from grain relationships in plutonic igneous rocks is a risky undertaking at best, as has been pointed out by many petrographers (Shand, 1947, pp. 105–116). If the rock has undergone recrystallization of some components, inversion in all major phases, and unmixing in a major constituent, it is futile to draw detailed conclusions concerning an order of crystallization from the mutual grain relationships. The texture of granitic rocks is partly mosaic and partly crystalloblastic, and apparently all gradations between the two are represented. Microcline and orthoclase in many granites appear to be the last minerals to crystallize; that is, they are molded around the plagioclase and quartz and appear to fill the interstices. This relation is reminiscent of a crystalloblastic fabric, and it is to be noted that orthoclase and microcline are at the bottom of the crystalloblastic series.

PERTHITE AND GRANITIZATION

The exsolution series from sanidine to perthite is convincingly demonstrated by the numerous descriptions of the various stages to be found throughout petrologic literature and by equilibrium studies of the alkali feldspars, which show that unmixing is demanded at low temperatures to maintain equilibrium. Perthites consisting of nearly equal amounts of albite and microcline, typified by those found in granites, such as the Quincy, Massachusetts, are prima facie evidence of magmatic ancestry. When

perthites of this type are present, granitization by any low-temperature process can be ruled out as improbable, if not impossible.

The presence of so-called "late-stage" albite at the Or-Pl boundaries indicates unmixing by solid diffusion and sets a limit beyond which diffusion in the solid state apparently did not operate. Indeed, the perthites themselves indicate the range over which solid diffusion was operative, a few millimeters at most, thus condemning solid diffusion to a minor role in the formation of these rocks.

METAMORPHISM

Metamorphism, particularly on a regional scale, will promote unmixing in the alkali feldspars. Larsen (1949) has described the metamorphism of sanidine-bearing volcanics under conditions corresponding to the green-schist facies of Eskola. The sanidine was unmixed to perthite. Where metamorphism was somewhat more intense, the sanidine unmixed to an irregular pattern of orthoclase and albite; and where the metamorphism was intense, the former presence of sanidine could not be detected, and it is assumed that complete unmixing[3] took place. Buddington (1939, p. 251 and *passim*) has described in detail the change from "primary" microperthite to a mixture of microcline and albite (or oligoclase) with increasing metamorphism of syenitic rocks.

The common occurrence of nearly pure albite and orthoclase, side by side, in granitic rocks suggests prolonged heating at relatively low temperatures as in regional metamorphism. Deformation may also play an important role in the unmixing process, as has been suggested by Phemister (1926, p. 25) and Buddington (1939, p. 251). Metamorphism is, then, capable of promoting the unmixing of alkali feldspars. The perthite to albite + orthoclase stage has been demonstrated by Buddington in the syenitic and granitic rocks of the Adirondacks and by Larsen in the southern California rocks. These field studies suggest that granites and syenites containing relatively pure albite and orthoclase are actually metamorphic rocks which formerly contained only one alkali feldspar. It is not certain whether the metamorphism responsible for the unmixing is the result of slow cooling (autometamorphism) or whether postintrusion regional and contact metamorphism have been operative. A careful study of the feldspars near contacts should provide much useful information.

SOME EVENTS IN THE COOLING HISTORY OF GRANITIC ROCKS

Granitic liquids low in lime will undoubtedly precipitate a single feldspar throughout the major part of primary crystallization. Laboratory studies indicate that volatile substances can probably lower the liquidus sufficiently to permit two feldspars to crystallize. But the amount of volatiles necessary to accomplish this is so great as to suggest that magmas rarely, if ever, contain enough to effect the crystallization, except, perhaps, during the final stages of consolidation after the bulk of the feldspar is already crystallized. On the other hand, from granitic magmas containing appreciable lime, two feldspars will crystallize in the early stages; but the plagioclase phase, as shown by the extru-

[3] The change from sanidine to a mosaic of albite and orthoclase has been called "recrystallization" by many investigators and has been used in the present paper in such a sense. However, if the phenocrysts went through the perthite stage or if the mosaic formed by grain-boundary unmixing, then the term "recrystallization" appears to be inadequate to describe the change.

sive rocks, will be considerably richer in anorthite than the liquid from which it is being precipitated, and the course of crystallization, as shown by Vogt (1926) and Larsen (1949), will cause the liquid to be depleted in lime. This removal of lime will result in the liquid's moving into the field of one feldspar, particularly if the plagioclase becomes zoned and fails to react with the liquid, as is so often seen in the extrusive rocks. Descriptions of feldspars in plutonic rocks indicate that cores of plagioclase surrounded by rims of perthite, cryptoperthite, or anorthoclase are common and suggest that such a course of crystallization has been followed.

During the final stage of crystallization the great concentration of volatile components may lower the liquidus enough so that orthoclase and plagioclase[4] would crystallize simultaneously in even lime-poor rocks. In such a case the alkali feldspar would show rims of plagioclase (Rapakivi?) on some grains and rims of a potash-rich phase on others if equilibrium between crystals and liquid is not attained.

During this stage the alkali feldspar will begin to unmix to perthite if the volatiles have lowered the liquidus to the appropriate temperature. If such a lowering has not taken place, the unmixing will occur at subsolidus temperatures and the exsolution will be metamorphic. Whether the reaction is autometamorphic or whether regional or contact metamorphism has been operative might be discovered by careful study of the feldspars near chilled contacts. Descriptions of the perthite-bearing rocks indicate that the anorthoclase to perthite stages can be autometamorphic. The perthite to orthoclase + plagioclase stage has been established as a metamorphic reaction, and further study is required to discover whether or not this change can take place during the period of initial cooling, i.e., can also be autometamorphic.

[4] It should be noted that, although two feldspars are precipitating simultaneously, the orthoclase would be rich in soda and the plagioclase rich in potash.

SUMMARY

Quartz and feldspars of the acidic plutonic rocks show much evidence of change from an initial character identical with the corresponding minerals of the extrusive equivalents. The quartz of plutonic rocks is different from the phenocrysts of the extrusives, as evidenced by inclusions, the high-low inversion, and petrofabric studies. Similarly, plagioclase feldspars show significant differences between plutonic and extrusive types. The alkali feldspars provide evidence of profound changes having taken place in the deep-seated environment of the slowly cooling granitic pluton.

Field and laboratory studies have shown that the feldspar of many acidic rocks has undergone unmixing from an originally homogeneous feldspar to an intergrowth of relatively pure potash and plagioclase feldspar. Unmixing has apparently taken place during initial cooling. Studies of metamorphism of these rocks have demonstrated that unmixing may be carried beyond the perthite stage and completely separate the two feldspars. The possibility that this process of complete unmixing may take place during primary cooling of plutonic rocks should be further investigated.

Heteromorphic pairs of granites, consisting of perthite and quartz, on the one hand, and microcline, plagioclase, and quartz, on the other, are particularly interesting, for they may represent complete unmixing of the feldspars.

Unmixing may be expected to be most important in the lime-poor acidic rocks, as the presence of lime permits two feldspars to coexist at higher temperatures. It is anticipated that little unmixing will be found in rocks such as the quartz diorites, where lime is an important constituent. On the other hand, syenitic rocks low in lime may be expected to exhibit considerable unmixing of the alkali feldspars.

In summary it may be said that the acidic plutonic rocks, as we now see them, may have undergone profound changes in mineralogy and textural relationships, and information as to the magnitude of the changes can be gathered only by studying in great detail the more quickly cooled contacts of such rocks.

APPENDIX

After the above manuscript was in proof, my attention was drawn to a paper by G. M. Schwartz (1931) in which he described experimental investigations of a sulfide solid solution which supports the theory that unmixing may continue beyond the perthite stage, resulting in complete separation of the two feldspars. This final stage in the exsolution series (perthite→ plagioclase + microcline) is difficult to establish experimentally because of the extreme sluggishness with which solid-state reactions proceed in silicates. However, the sulfides, where such reactions might be expected to proceed with greater facility, provide evidence for similar changes.

Schwartz has demonstrated experimentally that a solid solution of bornite-chalcopyrite will unmix, first, to an intergrowth analogous to perthite and, finally, to a granular aggregate of the two minerals (Schwartz, 1931, figs. 5, 6, p. 195). This complete separation of the two phases *takes place in 24 hours*. In discussing this unmixing, Schwartz (p. 196) writes:

> Unless the solid solution is drastically quenched, it breaks down and segregates with amazing rapidity into a crystallographic intergrowth of the subordinate mineral in the predominate one. A cooling time of five minutes from 600° C. to room temperature allows sufficient segregation so that the intergrowth may be observed at a magnification of 75 diameters. Cooling in one hour allows segregation of some of the subordinate mineral to grain boundaries, and 24 hours in some cases results in *complete segregation* into grains.

Again, on page 192, he writes:

> The complete segregation of the minor constituent into grains is most surprising and brings out a fact which has been very little considered in geologic theory; that is, *that even more or less granular mixtures of two constituents may represent a breakdown of a solid solution that existed at higher temperatures* [italics mine].

These results on sulfides are convincing evidence for the unmixing process which has been proposed for the alkali feldspars, and they emphasize the futility of using grain boundary relationships to determine an order of crystallization.

ACKNOWLEDGMENTS.—The writer is indebted to F. Chayes, of the Geophysical Laboratory, for many helpful suggestions made during the preparation of this paper and for the use of his large collection of thin-sections of granitic rocks. Thanks are especially due J. W. Grieg, of the Laboratory, and M. A. Ford, of the National Geographic Society, for assistance in preparing the photographs reproduced here.

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THE POLYMORPHISM OF $MgSiO_3$ AND SOLID-STATE EQUILIBRIA IN THE SYSTEM $MgSiO_3$-$CaMgSi_2O_6$[1]

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ABSTRACT

The investigation is divided into two parts: the first is concerned primarily with stability relations among the various crystalline phases of $MgSiO_3$. Contrary to results given in previous published investigations, experimental evidence favors the stability of rhombic enstatite and protoenstatite. Clinoenstatite is found to be a metastable low-temperature phase formed during rapid cooling of protoenstatite. The latter modification is the stable form above 990° C.; rhombic enstatite is stable below that temperature. A preliminary structure analysis of protoenstatite and clinoenstatite is presented. X-ray data lead to the conclusion that protoenstatite is also orthorhombic. Phase transitions appear to be attended by a one-dimensional stacking disorder in which intermediate states are unstable.

The second part of the investigation is concerned with subsolidus phase equilibria in the system $MgSiO_3$-$CaMgSiO_3$. Evidence is presented which corroborates Hess's findings that two monoclinic pyroxene phases may coexist in equilibrium. At lower temperatures a monoclinic diopside solid solution is in equilibrium successively with protoenstatite and rhombic enstatite solid solutions.

INTRODUCTION

The primary objective of this paper is to provide information about phase equilibria in the subsolidus system $CaMgSi_2O_6$-$MgSiO_3$. The latter compound is known to undergo polymorphic transitions; it is therefore necessary that the stabilities of the various $MgSiO_3$ forms and the inversion temperatures be known in order that a phase diagram for the binary system may be constructed. Considerable stress has been placed on this aspect of the problem because of the conflicting results given by previous authors. It has been found that at least a superficial knowledge of the various $MgSiO_3$ structures is a great aid in understanding the equilibrium relations among them; to this end, preliminary structure analyses have been performed on two of the $MgSiO_3$ phases. These analyses plus the known structure of rhombic enstatite have resulted in two hypothetical structures which serve to explain some of the apparently strange relationships in the $MgSiO_3$ system. It should be emphasized here that the crystallographic investigations undertaken in this investigation are not an end in themselves; they serve only to clarify the relations among the $MgSiO_3$ phases.

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THE POLYMORPHISM OF $MgSiO_3$

PREVIOUS INVESTIGATIONS

Perhaps the first thorough investigation of phase relations in the $MgSiO_3$ system is the classical work of Allen, Wright, and Clement (1906). These authors describe the synthesis of four crystalline modifications of $MgSiO_3$: an orthorhombic pyroxene (believed to be identical to natural rhombic enstatite), a monoclinic pyroxene (clinoenstatite), an orthorhombic amphibole, and a monoclinic amphibole. They concluded that the monoclinic pyroxene is the only phase which is stable at all temperatures and that the other phases crystallize metastably below about 1,100° C. A fifth crystalline phase of $MgSiO_3$ was

described by Allen, White, Wright, and Larsen (1909). This form, $\alpha MgSiO_3$, was obtained by heating clinoenstatite above 1,365° C.; the resulting material had orthorhombic symmetry and resembled forsterite in its habit and optical properties. However, Bowen and Anderson (1914) showed that $\alpha MgSiO_3$ is not the product of an inversion of clinoenstatite but is, instead, the result of dissociation of clinoenstatite to Mg_2SiO_4 (forsterite) plus a liquid.

The four original phases described by Allen, Wright, and Clement were accepted as the polymorphic forms of $MgSiO_3$ until Warren (1929) demonstrated that the only physically reasonable structure of tremolite that was compatible with observed X-ray data required the presence in the unit cell of 2.2 per cent of water by weight. Thus the "molecular" formula of tremolite was changed from $Ca_2Mg_6(SiO_3)_8$ to $H_2Ca_2Mg_5(SiO_3)_8$. In a later paper Warren (1930) compared the structures of several monoclinic amphiboles, among which was kupferrite, one of the amphibole phases described by Allen, Wright, and Clement. X-ray data revealed that all the amphibole structures were closely related and that all required the presence of water or fluorine in the unit cell. Thus the two amphibole phases could no longer be considered as polymorphic forms of $MgSiO_3$ because they actually had the stoichiometric composition $H_2Mg_7(SiO_3)_8$.

Meanwhile, in Germany, Haraldsen (1930), while investigating the decomposition of talc, $H_2Mg_3(SiO_3)_4$, to $MgSiO_3$ on heating, obtained a crystalline phase of $MgSiO_3$ that had an X-ray powder diagram different from those of rhombic and clinoenstatite. Haraldsen also succeeded in synthesizing the same phase by heating a stoichiometric mixture of MgO and SiO_2 to about 1,400° C. Apparently unaware of Haraldsen's work, Bowen and Schairer (1935) investigated the solid-phase equilibria of $MgSiO_3$ as part of a larger work on the system MgO-FeO-SiO_2. The latter authors concluded that both the rhombic and the monoclinic forms of $MgSiO_3$ are stable, even though they did not succeed in converting the monoclinic form to the orthorhombic form. The temperature of transition for pure $MgSiO_3$ was placed at 1,145° C.

The first investigation into the possible stability of Haraldsen's phase was performed by Büssem, Schusterius, and Stuckardt (1938), who found that the new modification, named "protoenstatite," possessed some rather remarkable properties: it converted to clinoenstatite on reheating to 1,300° or 1,400° C., at room temperature as the result of grinding, or even on long standing. The conclusion was that protoenstatite is unstable at all temperatures. Shortly thereafter, the $MgSiO_3$ system was re-examined by Thilo and Rogge (1939), who found that in the presence of LiF the transition rhombic enstatite-protoenstatite (renamed "mesoenstatite") was reversible in the region between 900° and 990° C. and that the transition mesoenstatite-clinoenstatite was also reversible, occurring at about 1,260° C. These results apparently provided considerable evidence in favor of the fact that there are three stable phases of $MgSiO_3$. Thilo and Rogge also described two other phases, called "M_1" and "M_2," formed by heating MgO and SiO_2 up to 1,000° C. and between 1,000° and 1,300° C., respectively, and also by the thermal decomposition of talc. These two phases were considered to be unstable at all temperatures. It is of interest to note at this point that the authors mention a

striking similarity in the X-ray powder diagrams of M_2 and clinoenstatite; there is also a similarity in the powder diagrams of M_1 and M_2, the former differing from the latter largely by having fewer lines.

The work of Thilo and Rogge appears to have been the last extensive published investigation of crystal-phase equilibria in the $MgSiO_3$ system. It is apparent from the foregoing discussion that there has been very little agreement regarding the number of crystal phases and the equilibrium relations among them. The first part of this paper, therefore, will be devoted to a re-examination of the system, as well as to an attempt to uncover the structural relations among the stable phases.

DESCRIPTION OF PHASES

The optical properties of rhombic and clinoenstatite have been described in several of the papers cited above. The values listed below are taken from Winchell (1931). The optical constants for protoenstatite were measured with sodium D light. Unit-cell parameters for all three crystalline forms were calculated from X-ray powder photographs taken in a Norelco camera of 114.59-mm. diameter. Although the camera employs the Straumanis film mount, an independent calibration was performed with NaCl. The required powder lines were indexed with the aid of unit-cell values taken from the literature and, in the case of protoenstatite, by means of data obtained from c-axis rotation photographs. The d spacings listed in table 1, were calculated from the same powder films as those from which the unit-cell parameters were taken.

RHOMBIC ENSTATITE

Rhombic enstatite appears under the microscope as lath-shaped crystals showing parallel extinction. Cleavage on (110) at about 88° is characteristic of all three polymorphic forms. Twinning is rare after (104) and (011).[3] The latter twin law will be discussed in greater detail in the section on structural relations. The optic plane is (010); $Z = c$; +; $2V = 60°$;[4] $\gamma = 1.658$; $\beta = 1.653$; $\alpha = 1.650$; $\gamma - \alpha = 0.008$. The unit translations are: $a = 18.20 \pm 0.05$ Å; $b = 8.89 \pm 0.05$ Å; $c = 5.20 \pm 0.05$ Å ($a = 18.20$ Å; $b = 8.86$ Å; $c = 5.20$ Å, Warren and Modell, 1930). The d spacings measured for the synthetic material (table 1) compare favorably with the values published by Clark (1946) for the natural mineral; the writer's d values average between 0.005 Å and 0.01 Å higher than those listed by Clark.

PROTOENSTATITE

Crystals of protoenstatite suitable for optical measurements under the microscope were prepared by heating finely powdered natural enstatite plus a few per cent of LiF at about 1,100° C. for a week. An X-ray powder diagram of the resulting material proved that it had been converted to protoenstatite. It is important to note at this point that samples for X-ray powder work were made by breaking a small elongate sliver off the fused polycrystalline cake resulting from the heat treatment. Crushing the cake to form a rod by orthodox methods would have resulted in conversion of the material to clinoenstatite.

Protoenstatite synthesized as described above appears as lath-shaped

[3] These indices are commonly listed as (014) and (101). The a- and b-axes have been interchanged here to conform to the usage of Warren and Modell.

[4] This value was obtained from Winchell (1931, p. 295, fig. 271). Winchell's recorded value of $2V = 31°$ was apparently taken from Allen, Wright, and Clement (1906). The recorded value contradicts the one given in the figure.

crystals, having parallel extinction, which apparently are pseudomorphic after the low form. As in rhombic enstatite, Z is parallel to the longest dimension of the crystals, $=c$. As far as could be determined, the indices differ no more from those of the unheated mineral[5] than the experimental error, about ± 0.002. The value of $2V$ is large, about 70°, optical sign +. In the light of these facts it is now readily understandable why the proto-form remained undiscovered until the application of X-ray powder methods.

Evidence favors the following unit translations: $a = 9.25$ Å; $b = 8.92$ Å; $c = 5.25$ Å, all values ± 0.05 Å. The d spacings in table 1 compare favorably with those of Clark, averaging, as before, about 0.01 Å larger than the values listed by that author.

CLINOENSTATITE

Clinoenstatite commonly appears as prismatic crystals or as tablets after (100). Polysynthetic twinning after (100) is characteristic. The optic plane is normal to (010). The data are as follows: Z lies in the obtuse angle β; $Z \wedge c = 22°$; $2V = 53\frac{1}{2}°$ for pure $MgSiO_3$ as clinoenstatite; +; $\gamma = 1.660$; $\beta = 1.654$ (calculated); $\alpha = 1.651 \pm 0.003$; $\gamma - \alpha = 0.009$.

The unit translations are: $a = 9.12$ Å; $b = 8.86$ Å; $c = 5.24$ Å ± 0.05 Å; β, as determined from a precession photograph, is $92°45' \pm 0'.10$ ($\beta = 92°24'$, Winchell). The measured d spacings are presented in table 1.

STRUCTURAL RELATIONS

RHOMBIC AND CLINOENSTATITE

The structure of rhombic enstatite (fig. 1, *A*), deduced by Warren and Modell (1930), forms the foundation upon which the following discussion rests. By means of this structure and certain additional data to be presented, reasonable structures for the remaining two $MgSiO_3$ phases may be determined. The space group of rhombic enstatite is V_h^{15}, or *Pbca;* the unit translations have been given in the section on the description of phases. The unit cell of rhombic enstatite in figure 1, *A*, may be considered to be composed of two monoclinic cells, each having $a = 9.10$ Å, joined by the b-glide plane normal to a. The angle β of these monoclinic cells may be readily calculated: Warren and Modell give the Z co-ordinate of Si_2 as 0.04. It is apparent, then, from figure 1, *A*, that $Z(Si_2) - Z(Si_2') = 0.08$. The absolute distance is then 0.08×5.20 Å, or 0.416 Å. The x distance between the two Si atoms is:

$$(9.1 \text{ Å} - 0.06 \times 18.20) = 8.01 \text{ Å};$$

therefore $0.416/8.01 = \tan(\beta - 90°)$. Solution of this equation gives $\beta = 92°57'$, which is in good agreement with the measured value of 92°45′ for clinoenstatite. It will be shown later that rhombic enstatite and clinoenstatite are separated by a reconstructive transition and a displacive transition. However, the close correspondence between the computed angle β for the monoclinic subcells of rhombic enstatite and the measured angle for clinoenstatite indicate a similarity between the two structures. A hypothetical structure for clinoenstatite is shown in figure 1, *B*. Precession photographs show that clinoenstatite has an a-translation of 9.12 and that the other two translations are the same as those of rhombic enstatite.

The extinction pattern reveals that the unit cell is primitive and that the glide plane (010) with component $c/2$ is present. The extinction of ($hk0$) reflec-

[5] The natural enstatite is from Bamle, Norway. Analysis by optical methods showed that it contained less than 5 mol per cent of $FeSiO_3$.

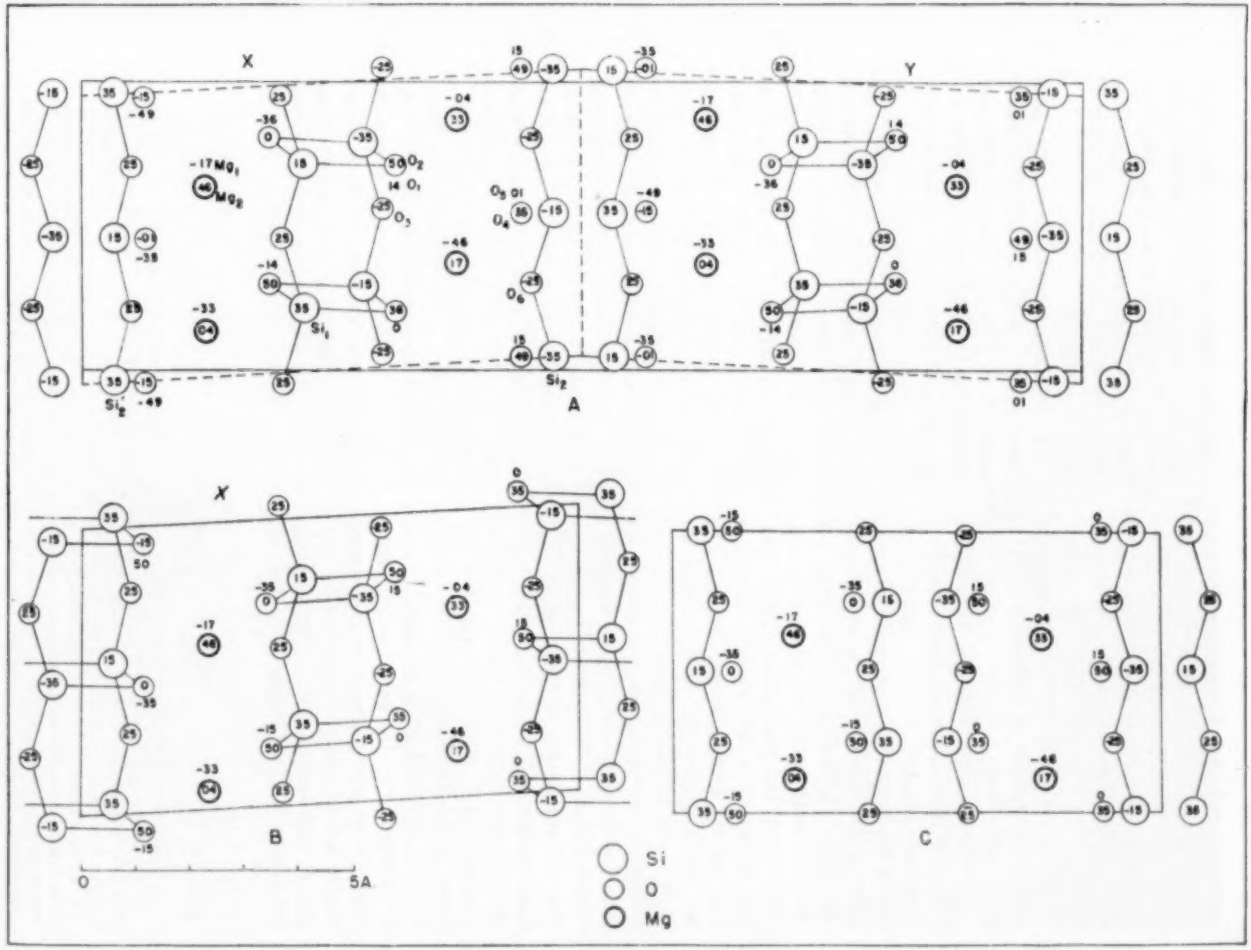


FIG. 1.—*A*, The structure of rhombic enstatite, after Warren and Modell. *B*, Hypothetical structure of clinoenstatite. *C*, Hypothetical structure of protoenstatite. The *c*-axes are vertical, *a*-axes horizontal, and *b*-axes normal to the page. Numbers give approximate *Y* co-ordinates of atoms in decimal parts of the *b*-translation distance.

tions when $h + k$ is odd implies that the projection of the structure on the (001) face is centered. This projected c-centering also occurs in rhombic enstatite, causing the extinction of (hk0) spots when $h/2 + k$ is odd. The (hk0) extinctions due to projected centering mask the extinction pattern of a possible twofold screw axis [010].

If the transition from one form to the other entails motion of the atoms in a direction parallel to the c-axis only, then whatever (hk0) spots the two forms have in common should have the same intensities. In the transition from the rhombic form to the hypothetical monoclinic form, the glide plane (100), causing extinction in hk0 where h is odd, is lost, but the a-translation is halved. The net result is that the (hk0) pattern for both forms should be identical. This is indeed the case; rotation photographs for both forms have 0 layer lines which are identical (pl. 1). The above facts show that clinoenstatite has the same unit translations and angle β as the hypothetical structure and that clinoenstatite may be derived from the rhombic form by a displacement of the atoms entirely, or almost entirely, parallel to c, as is required by the hypothetical structure. Other structures may be drawn which satisfy the restrictions discussed above; however, some of these other structures involve highly distorted co-ordination shells about the Mg atoms, and others require four very different Mg positions per unit cell. The structure shown has co-ordination shells about the Mg atoms which are similar to those in rhombic enstatite.

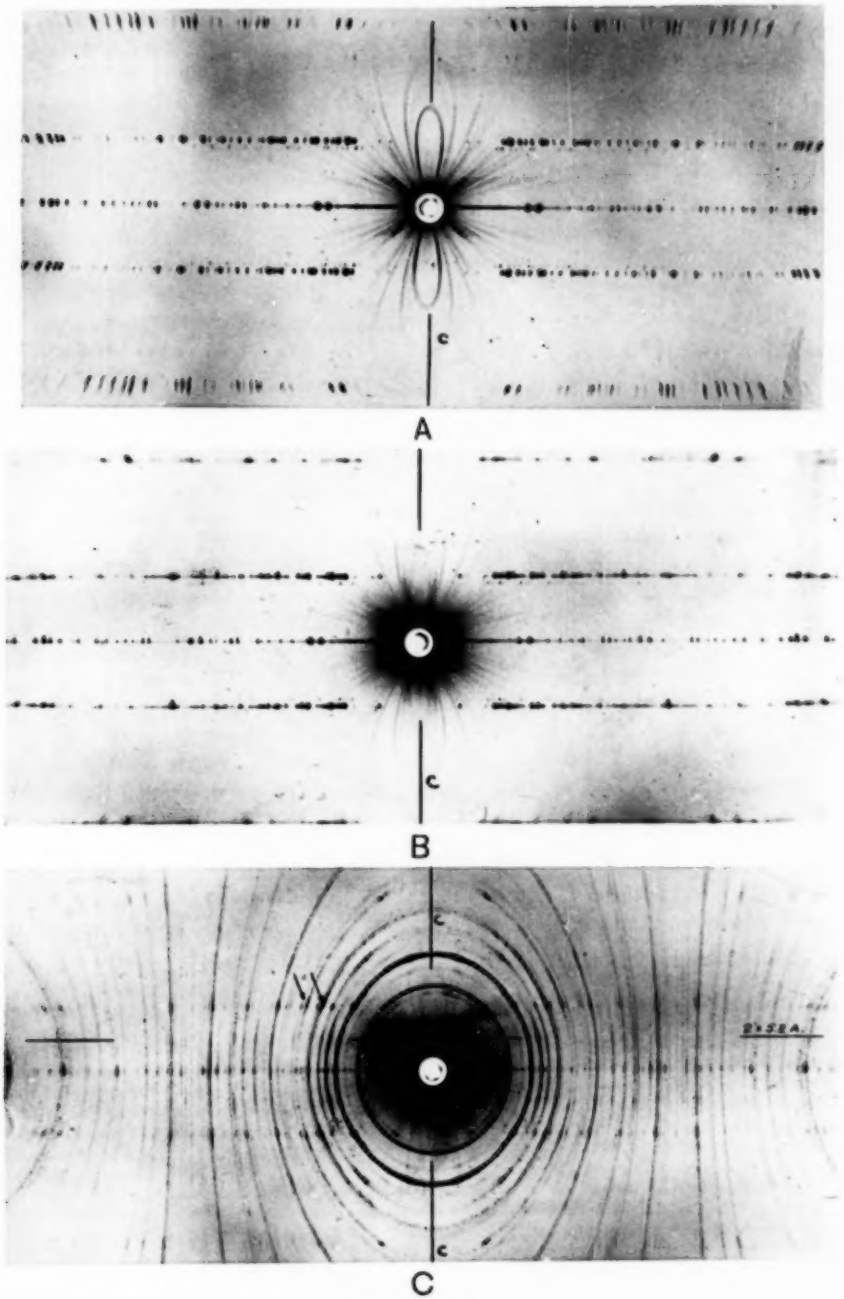
Clinoenstatite appears as the final phase when a single crystal of rhombic enstatite is heated above 1,200° and quenched in air. The stages of this transition were studied by means of 0-level, b-axis precession photographs (pl. 2) taken of rhombic enstatite which had been heated to various temperatures for different times and quenched. Plate 2, *A*, is the pattern given by unheated natural enstatite; 2, *B*, by a crystal heated at 1,250° C. for 16 hours; and 2, *C*, by another crystal heated at 1,400° C. for 20 hours. For the purpose of the following discussion, one of the two monoclinic cells forming the unit cell of rhombic enstatite will be designated by the letter X, the one having the other orientation by the letter Y. Thus Warren and Modell's structure may be represented as: XYXYXYXY . . . , whereas clinoenstatite is given by *XXXXXX* . . . or *YYYYY* . . . (*X* represents a modified X cell, as shown in fig. 1, *B*). Consideration of the relations between the monoclinic and rhombic structures shows that a vast number of states intermediate between the two end-structures is possible. Thus XYXYX*Y*YXYXY . . .

PLATE 1

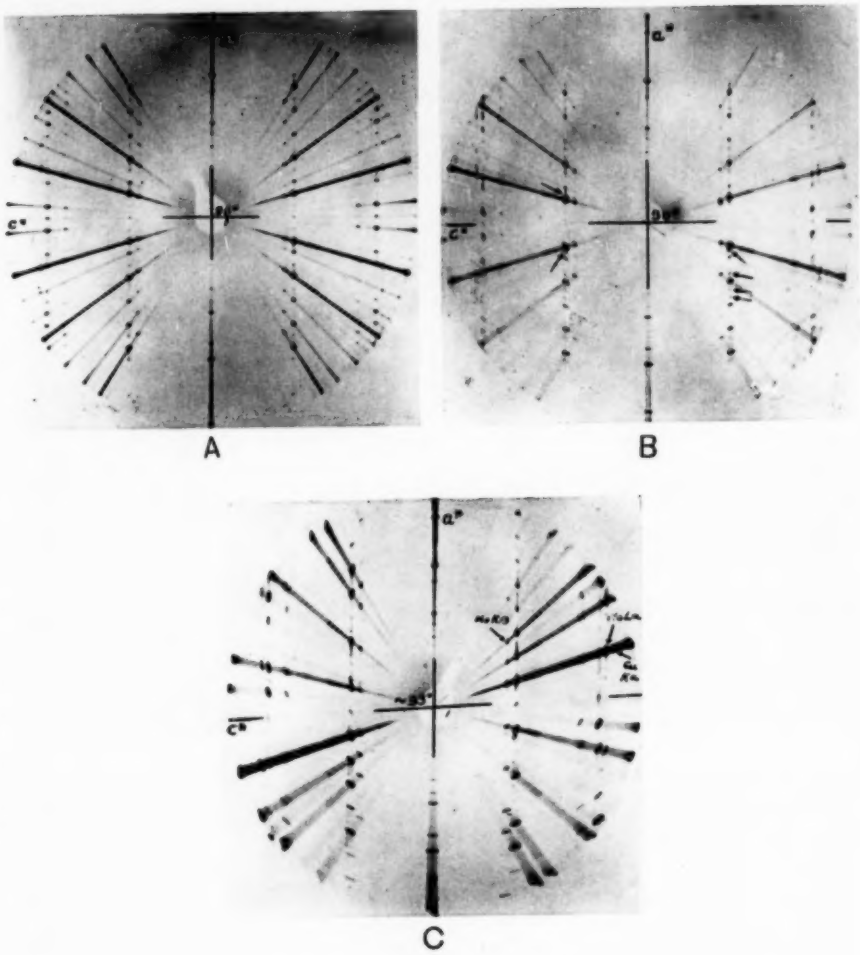
A, Unheated rhombic enstatite from Bamle, Norway; rotation about the c-axis.

B, Clinoenstatite prepared as described in the text; rotation about the c-axis. The diffuse streaks along the n layer lines are caused by a random orientation of unit cells along the a-axis. Note that the 0 layer lines of *A* and *B* are nearly identical and that they are similar to that of *C*.

C, Protoenstatite obtained by heating fibers of anthophyllite as described in the text. The c-axis is parallel to the longest dimension of the fibers; no rotation was necessary. Note that the spots of 10.3 Å layer line are greatly drawn out along powder arcs. The arrows show that the arcs of the layer lines corresponding to submultiples of that spacing lie slightly outside the spots of the 5.2 Å layer lines. All three photographs were taken with Fe radiation filtered through Mn. The radius of the camera is 30.0 mm.; the photographs are approximately five-sixths the size of the original films.



X-ray photographs of enstatites



X-ray photographs of enstatites

would closely resemble rhombic enstatite, whereas $(XXX)_n(YYY)_m$ would be clinoenstatite twinned after (100), if n and m are fairly large. It is readily apparent that in the intermediate states a regular identity period may extend only very short distances parallel to the a-axis. On a precession photograph this lack of periodicity appears as diffuse streaks between spots having the same l-index. In order to show that the diffusion was indeed parallel to the a-axis, i.e., that there was no component of the diffusion parallel to b, a-axis precession photographs were taken. These showed no trace of diffusion whatsoever.

Plate 2, *A*, shows the presence of diffuse streaks, demonstrating the existence of randomness in even the natural mineral. This is of interest in the light of Zambonini's (1908) and Ito's (1935) contentions that rhombic enstatite may not have a perfectly regular structure but may be considered as clinoenstatite *statistically* twinned on a unit-cell scale. It will be noted that, although the spots of plate 2, *A*, have orthorhombic symmetry, the diffuse streaks have monoclinic symmetry, and they are associated with every other rhombic spot. These diffuse streaks are the first evidences of the monoclinic spots which will appear after the transition has proceeded to a greater degree. It is remarkable that the monoclinic diffuse streak pattern indicates a lack of twinning after (100).

Plate 2, *B*, represents a later stage in the conversion of rhombic to clinoenstatite. Diffuse streaks parallel to a are still present, but definite spots forming a monoclinic pattern have appeared. The monoclinic pattern is here somewhat masked by the fact that it is twinned after (100). The orthorhombic pattern is still present. In this photograph and in plate 2, *C*, the radial streaks have become broadened so that they cover an arc of a degree or two. This is a result of the development of flaws and cracks in the crystal after heat treatment and quenching.

Plate 2, *C*, shows almost complete conversion to clinoenstatite. The spots of the orthorhombic pattern have in almost every case vanished, spreading out into diffuse streaks. The situation is now the reverse of plate 2, *A;* the spots form a monoclinic pattern, which again is untwinned, whereas the streaks form an orthorhombic pattern. The angle β is now equal to 92°45′, and $a = 9.1$ Å.

Evidence will be presented in a later section that rhombic enstatite passes through the protoenstatite structure during the transition to clinoenstatite. This indicates that the first stage of the transition rhombic enstatite → clinoenstatite involves the formation of proto-

PLATE 2

A, Unheated rhombic enstatite; the b-axis is the precession axis, 0 level. Note that the spot pattern indicates the presence of planes of symmetry normal to a^* and c^*. The diffuse streak pattern, however, is based on a twofold axis parallel to b^*, with no symmetry planes normal to a^* and c^* and no twofold axis parallel to a^* and c^*. The diffuse streaks connect every other pair of spots, suggesting a halving of the a-translation.

B, Rhombic enstatite heated to 1,250° C. for 16 hours; b-axis precession, 0 level. The arrows indicate a new series of spots forming a twinned monoclinic pattern. The spots corresponding to the orthorhombic patterns are dominant.

C, Rhombic enstatite heated to 1,400° C. for 20 hours; b-axis precession, 0 level. A monoclinic spot pattern is now dominant. The diffuse streaks and a few faint spots still form an orthorhombic pattern. The a-translation is now half the value of photograph *A*. All three photographs were taken with unfiltered Mo radiation. The target of the Mo tube contained traces of W and Cu contamination.

TABLE 1

POWDER X-RAY DATA FOR $MgSiO_3$ PHASES

NATURAL RHOMBIC ENSTATITE			SYNTHETIC RHOMBIC ENSTATITE*			SYNTHETIC PROTOENSTATITE†			SYNTHETIC CLINOENSTATITE‡		
d(Å)	I	*hkl*	*d*(Å)	I	*hkl*	*d*(Å)	I	*hkl*	*d*(Å)	I	*hkl*
9.41	8										
7.36	1										
6.32	1	(210)	6.30	1	(210)	6.31	1	(110)	6.32	1	(110)
4.55	7	(400)									
4.40	4		4.38	2		4.36	2		4.40	3	
									4.28	1	
			4.02	3		4.07	1		4.04	1	
3.61	1										
3.46	1					3.48	1		3.49	1	
3.28	5		3.28	3		3.23	2		3.27	5	
3.14	10	(420)	3.14	10	(420)	3.16	10	(220)	3.16	7	(220)
2.92	4		2.92	4					2.97	7	
2.85	10	(610)	2.85	6	(610)	2.89	6	(310)	2.86	10	(310)
2.81	2		2.81	2							
2.69	4		2.69	4		2.72	4				
						2.66	5				
2.52	7		2.52	4					2.53	3	
2.48	6		2.48	6					2.51	2	
2.46	6		2.46	3		2.45	3		2.45	4	
									2.42	3	
			2.37	1					2.37	2	
2.35	1		2.35	1							
2.27	1					2.29	4		2.27	1	
2.24	1		2.24	1							
2.22	1	(040)	2.22	1	(040)	2.22	3	(040)	2.20	3	(040)
2.10	5	(630)	2.10	5	(630)	2.11	2	(330)	2.11	6	(330)
2.08	5		2.08	4					2.09	1	
2.05	3		2.05	3		2.06	2				
2.02	2		2.02	2					2.02	2	
1.98	4		1.98	3		1.97	7		1.98	1	
1.95	4		1.95	4					1.96	1	
			1.92	1					1.92	1	
1.88	2		1.88	1							
1.85	1		1.85	1					1.84	1	
1.83	1		1.83	1		1.83	1				
1.783	3	(10.10)	1.782	2	(10.10)	1.809	2	(510)	1.784	3	(510)
1.769	2		1.770	1							
1.728	4		1.730	2		1.742	1		1.730	2	
1.697	2		1.698	2		1.715	4				
1.678	1		1.676	2		1.687	1				
			1.648	1		1.658	1				
			1.640	1		1.640	5		1.640	2	
			1.628	1		1.621	2				
1.604	3		1.606	3					1.606	6	
1.586	4		1.585	2		1.586	1		1.585	1	
1.524	7		1.526	1		1.523	1		1.526	4	
			1.517	2					1.520	4	

* Synthetic rhombic enstatite used for this determination was prepared by heating a stoichiometric oxide mixture plus about 2 per cent of LiF at 950° C. for 2 days.

† This sample of protoenstatite was prepared by heating $MgSiO_3$ glass at 1,300° C. for 1 day without a flux.

‡ Clinoenstatite for this determination was obtained by heating the oxides plus LiF at 1,200° C. for 2 days.

TABLE 1—*Continued*

Natural Rhombic Enstatite			Synthetic Rhombic Enstatite*			Synthetic Protoenstatite†			Synthetic Clinoenstatite‡		
d(Å)	I	*hkl*	*d*(Å)	I	*hkl*	*d*(Å)	I	*hkl*	*d*(Å)	I	*hkl*
1.482	7	(060)	1.484	8	(060)	1.495	7	(060)	1.482	1	
1.468	6		1.471	4		1.456	3		1.469	5	
1.418	1		1.417	2							
1.391	5		1.395	2		1.404	1		1.397	1	
			1.389	4							
1.378	2		1.377	2		1.375	3		1.375	7	
1.359	1		1.359	1		1.360	3		1.356	1	
1.338	1		1.326	2		1.334	1		1.337	1	
									1.318	2	
1.306	3	(004)	1.300	3	(004)	1.314	6	(004)	1.310	1	(004)
1.297	2		1.290	3							
1.288	2		1.288	3					1.283	2	
			1.273	1		1.272	1				
1.266	4		1.262	4		1.261	4		1.264	3	
1.252	2		1.246	1					1.257	3	
			1.240	1					1.239	1	
1.229	2		1.223	1		1.227	5				
1.208	2		1.202	2					1.209		
1.184	2		1.178	2							
1.162	1					1.167	1		1.162	1	
1.140	1					1.143	3		1.136	1	
1.126	1		1.122	1							
1.116	1					1.116					
1.104			1.101	1					1.101	2	
1.082	3		1.079	2		1.091	1				
			1.077	2							
1.071	2		1.070	1					1.065	1	
1.061	2		1.060	2		1.061	2		1.062	3	
1.057	2		1.055	1		1.059	3		1.055	1	
1.052	2		1.050	1		1.052	4				
1.048	5		1.046	5		1.041	2		1.047	4	
1.037	2		1.035	1					1.035	1	
			1.033	2		1.033	1		1.030	2	
1.027	1		1.027	1							
1.021	3		1.019	5							
1.012	4		1.011	5		1.007	2		1.011	2	
0.994	1		0.992	1		0.995	5		0.998	2	
0.989	2		0.988	1					0.988	1	
0.985	3		0.984	4							
0.980	3		0.980	5					0.979	1	
0.974	3					0.976	4		0.975	1	

enstatite unit cells distributed in a random way throughout the heated crystal. These protoenstatite unit cells then transform to monoclinic cells during the quench or by further reaction at the high temperature (evidence supports the former mechanism). The monoclinic cells thus formed are arranged in more or less random fashion. The longer the time of heating and the higher the temperature, the more complete is the conversion of rhombic enstatite to protoenstatite and finally to clinoenstatite. Under the microscope, heated crystals of rhombic enstatite exhibit a few broad twin bands separated by very diffuse boundaries. Extinction is wavy, and $Z \wedge c$ is generally less than 22°.

It is clear that, when only a few protoenstatite cells are formed during the heat

treatment, the crystal will still greatly resemble rhombic enstatite in its X-ray patterns, even after the protoenstatite cells have converted to monoclinic cells.

The greater the number of protoenstatite and, finally, clinoenstatite cells produced, the greater is the possibility of resemblance of the X-ray pattern of the heated material to that of clinoenstatite. The orientation of the monoclinic cells formed from the protoenstatite cells will be a function largely of the distribution of stresses acting on the surfaces of the crystal and on discontinuities within it and on the influence of the orientation of nearest neighbors upon a given cell. The tendency is for the newly formed monoclinic cells to be arranged in the pattern *XXXXX* . . . rather than being reconverted to the regular pattern XYXYXY . . . . The reason for this will be shown to be the fact that much larger atomic displacements from the protoenstatite structure must occur to regain the (100) glide plane in the pattern XYXYXY . . . than to form the pattern *XXXXX*. . . .

PROTOENSTATITE

In this section an attempt will be made to present some information about the structure of protoenstatite. Unfortunately, the writer was unable to synthesize a crystal of that phase of sufficient size and perfection for precession camera work. Therefore, the only photographs available for structure determination are *c*-axis fiber photographs. The fibers of the proto-form were prepared in the manner described by Thilo and Rogge (1939, p. 360); anthophyllite fibers were heated at about 1,250° for 16 hours. A typical picture of this material appears in plate 1, *A*. Certain crystal-chemical evidence to be cited in detail below compensates in some small measure for the lack of X-ray data.

Unit translations.—Examination of table 1 indicates that the major differences in the powder patterns of the three crystalline forms lie in the number of lines present and their intensities rather than in their spacings. It is thus evident that all three forms are based on approximately the same unit translations or integral multiples or submultiples thereof. Thilo and Rogge (1930, p. 362) published the value $c = 10.48$ Å for protoenstatite. This value was based on the appearance of a weak layer line corresponding to that distance on a *c*-axis fiber photograph. An alternate explanation for that layer line is possible. It is conceivable that the lattice of protoenstatite may have one or more rational directions along which, by coincidence, the identity period is double that of the *c*-axis. If such is the case, then it is also possible that the heated fiber may contain crystallites having two or more preferred orientations, parallel to the fiber axis, one of which is *c*, the others being the rational directions mentioned above. This situation would also result in a layer line corresponding to about 10.50 Å, even though *c* is 5.25 Å. If, for the moment, we assume that $b = 8.92$ and $c = 5.25$ Å, and not simple multiples of those values, then it is found that the identity period of the direction [011] is 10.35 Å. On the other hand, if we allow *a* to be 9.25 Å, then the direction [101] has the translation distance 10.62 Å when $c = 5.25$. By comparison, $2c = 10.50$ Å. Evidence to be discussed below favors the hypothesis of preferred orientations with $c = 5.25$. Moreover, one of the preferred orientations is [011]. Careful examination of plate 1, *B*, reveals that the second layer line (the first layer line of $c = 5.25$) contains some spots which lie slightly but definitely farther out than the others. The line of the outer

spots corresponds to an identity period of 10.3 Å, which is in good agreement with the identity period along [011]. Additional evidence in favor of preferred orientations is listed below:

a) Fiber photographs taken of heated anthophyllite which had been slowly cooled (over a period of 1 day) displayed the pattern of protoenstatite. Nevertheless, the spots of the 10.3 Å layer lines were so drawn out along powder arcs that those layer lines could no longer be said to exist. However, the spots of the 5.2 Å layer lines had become much sharper.

b) A layer line corresponding to a spacing of 10.3 Å may be observed on some rotation pictures of rhombic enstatite formed by heating anthophyllite fibers at 1,050° C. for a day. It can readily be shown that preferred crystallite orientations such that [001] and [011] are parallel correspond approximately to the twin law (011). Under the microscope some heated crystals of rhombic enstatite show intergrowths at angles of approximately 60°, corresponding to the above twin law.

The foregoing discussion has shown that b cannot have a value equal to $n\times$ 8.92 Å (where n is an integer), because there would then be no rational direction in the lattice having an identity period of 10.3 Å. Several reflections on the first layer line of the *c*-axis fiber photograph shown in plate 2, *C*, were indexed, using unit translations $a =$ 18.50 and $b = 8.92$ Å. At least one unequivocally indexed spot was found corresponding to an odd value of k. This eliminates the possibility that $b =$ 8.92/2. Moreover, (hkl) reflections were found with indices such that the sums $h + k + l$, $h + k$, $k + l$, and $h + l$ were odd when a was assumed to be either 18.50 or 9.25 Å. This indicates that the unit cell is primitive for both values of a.

The value of the a-translation could not be definitely established with the available X-ray data. There is, however, some evidence in favor of the value 9.25 Å over 18.50 Å. All the possible indices for about twenty (hkl) reflections in the first and second layer lines were determined on the basis of $a = 18.50$ Å. It was found that in most cases no reflection appeared corresponding to an odd h-index. Where such a reflection was present, one or more other indices having h even were equally possible. Several unequivocally indexed reflections with odd h-indices, whose counterparts were missing on the protoenstatite fiber photograph, were present on *c*-axis rotation pictures of rhombic enstatite. The further possibility exists that some of the spots considered to be absent were actually present but were so weak that they were not apparent above the confused pattern of powder arcs on the fiber picture. This possibility might be expected if the unit cell does have $a =$ 18.50 but if the two halves of the cell are very similar, differing only, for example, in the Z co-ordinates of the Mg atoms.

Symmetry and structure.—Keeping in mind the limitations on the reliability of indices and the uncertainty of establishing true extinctions, other extinction laws may be tentatively determined. The film co-ordinates of about sixteen ($0kl$) reflections, sixteen ($h0l$) reflections (based on $a = 9.25$ Å), and seventeen ($hk0$) reflections were computed. It was found that, for ($0kl$), spots were present only if k was even, and for ($hk0$) only if the sum $h + k$ was even; ($h01$) reflections were absent, but several ($h02$) spots were found. Wherever spots appeared in contradiction to the above extinction patterns, other allowable in-

dices were equally possible. If these extinction laws are assumed to be correct, it is evident that the protoenstatite lattice contains the glide planes (010) and (100) with components $c/2$ and $b/2$, respectively. The ($hk0$) extinctions may be caused by the glide plane (001) with component $a/2 + b/2$ or, more generally, by the fact that the projection of the structure on the (001) face may be centered.

The above data plus the parallel extinction noted previously suggest that protoenstatite has orthorhombic symmetry. If the above extinction patterns are accepted, the minimum symmetry is given by *Pbc*2. (The usual orientation is written as *Pca*.) The space group *Pbcn* represents the maximum symmetry allowable.

Comparison of the 0 layer lines of a fiber photograph of protoenstatite and a *c*-axis rotation photograph of rhombic or clinoenstatite reveals a great similarity except for slight displacements of some protoenstatite reflections toward lower-glancing angles. These displacements are explained readily by the larger *a*-value characteristic of that phase. The above facts indicate that the protoenstatite should be derivable from either of the other two structures by atomic displacements which are for the most part, or entirely, parallel to the *c*-axis. Although a determination of the structure amplitude is beyond the scope of this paper, a certain amount of information may be obtained from a qualitative consideration of the intensities of the (004) plane in the three phases. It will be noted that the intensity of (004) in protoenstatite is higher than that of the same plane in rhombic and clinoenstatite.

A hypothetical structure for protoenstatite is shown in figure 1, *C*. This structure has the space-group symmetry *Pbc*2, and it is derivable from the rhombic enstatite structure by atomic displacements which are for the most part parallel to *c*. The high intensity of (004) is explained by the fact that in the protoenstatite structure all the Si and O atoms lie in layers normal to [001], which are separated by one-fourth the *c*-translation distance. Thus, in the case of the (004) reflection, the amplitudes of waves from atoms in these layers reinforce one another. This situation does not occur exactly in the other two structures, where the chains are staggered.

Two of the crystal-chemical properties of protoenstatite provide considerable information relative to its structure: they are the ready conversion to clinoenstatite on grinding and the elimination of the protoenstatite stability field with substitution of Ca for Mg in its structure. The conversion of protoenstatite to clinoenstatite as a result of grinding or long standing at room temperature indicates that both structures must be very similar. Indeed, this transition evidently requires a lower activation energy than does the transition rhombic → clinoenstatite; even long-continued grinding of rhombic enstatite does not yield clinoenstatite. Consideration of figure 1, *C*, reveals that it requires smaller atomic displacements parallel to the *c*-axis to convert two protoenstatite unit cells into two clinoenstatite unit cells than it does to transform one rhombic enstatite cell into two clinoenstatite cells. Grinding protoenstatite results in the random application of stresses parallel to the *c*-axes of the tiny crystals in the powder, with the consequent formation of monoclinic cells by simple displacement. Under the microscope many of the clinoenstatite crystals formed in this way exhibit blurred twin boundaries and wavy extinction, clearly showing a high degree of randomness along the *a*-axis.

Büssem, Schusterius, and Stuckardt (1938) and Weyl (1949) have remarked that protoenstatite formed from a glass or in the presence of a material which yields a glass in the cooled sample and appears to have greater stability at room temperature, the increase in stability being caused by surface effects or by the protective action of glassy envelopes about the protoenstatite crystals.

At temperatures slightly above 1,350° C., $MgSiO_3$ as clinoenstatite forms a complete solid-solution series with the compound $CaMg(SiO_3)_2$ (diopside). Diopside is monoclinic with $\beta = 105°50'$. The intermediate members of this solid-solution series have angles which are a function of the percentage of Ca in the structure; an increase in the amount of Ca present increases the angle β. It will be shown in the second part of this paper that Ca is less soluble in protoenstatite than in the other two phases. It is evident that Ca introduces a *c*-axis shear in the $MgSiO_3$ structure, thus increasing the angle β. Protoenstatite apparently has a structure representing the least shear parallel to *c* of the three polymorphic forms; hence the introduction of Ca will tend to convert it into the more highly sheared rhombic or monoclinic structures.

In summary, atomic positions in the protoenstatite structure are apparently between those in the rhombic enstatite and clinoenstatite structures. Unstable states having more or less random distributions of unit cells along the *a*-axis exist between any two of the three phases. It is very likely that the M_1 and M_2 forms described by Thilo and Rogge represent such random states.

PHASE EQUILIBRIA

PROCEDURE

Two or 3 mg. of finely powdered starting material, enclosed in platinum-foil envelopes, were heated in platinum-, kanthal-, or nichrome-wound furnaces. Temperature control to $\pm 5°$ C. was provided by Leeds and Northrup Micromax and Tagliabue Celect Ray controllers. A continuous temperature record was obtained on a Brown eight-point recorder.

It was found necessary to utilize various fluxes in order to accelerate the reaction of runs starting with or yielding rhombic enstatite. About 2 per cent by weight of LiF was found to be the most effective of these substances. The use of fluxes has provoked considerable controversy regarding the possibility that they may have an effect not only on the velocity of a reaction but also on its equilibrium point; therefore, runs were made about the suspected inversion points with several fluxes. At the termination of a run, samples were air-quenched. Starting materials consisted of $MgSiO_3$ glass, natural enstatite from Bamle, Norway, stoichiometric mixtures of MgO and ground SiO_2 glass, and various phases of $MgSiO_3$ synthesized from the above materials.

The final phases resulting from heat treatment were determined by X-ray powder methods according to the technique discussed under the description of protoenstatite.

EXPERIMENTAL RESULTS

Thermal results are presented in table 2. If only the nonitalicized runs of table 2 are considered, the equilibrium relations among the three phases appear to resemble the pattern suggested by Thilo and Rogge; the only difference lies in the temperatures at which the inversions occur. The nonitalicized runs suggest two inversion points: rhombic enstatite $\rightleftarrows$ protoenstatite at 990°, and protoenstatite $\rightleftarrows$ clinoenstatite at 1,160 C. Thilo and Rogge place the first point be-

tween 900° and 990° C. and the second at about 1,260° C. Explanations for the anomalous italicized runs are possible, using the above equilibrium scheme. The presence of protoenstatite in runs above 1,160° may be caused by unstable crystallization of the glass as protoenstatite, with the conversion of that phase to clinoenstatite being prevented by insufficient reaction time. This hypothesis will explain the higher value of the second inversion point obtained by Thilo and Rogge; their runs lasted a few hours, and no reverse runs below 1,270° were made. The presence of clinoenstatite in runs below 1,160° might be explained by transformation of the protoenstatite by the thermal stresses set up by a quench. Büssem, Schusterius, and Stuckardt mention that better preservation of protoenstatite is obtained if a run is annealed somewhat. This has not been observed by the writer.

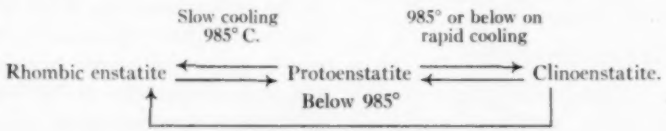
TABLE 2

THERMAL DATA FOR THE POLYMORPHISM OF $MgSiO_3$

Initial Material*	Flux	Time (Days)	Temp. (° C.)	Final Phases
Pr En	NaF	9	975	Pr En+R En
Nat En	LiF	8	980	R En
En gl	LiF	8	980	Pr En+R En
Cl En	LiF	8	980	R En+Cl En
Nat En	LiF	5	995	R En+Pr En
En gl	LiF	5	995	Pr En+Cl En
En ox	LiF	5	995	Pr En+Cl En
Cl En	LiF	5	995	Pr En+Cl En
Nat En	LiF	7	1,110	Pr En+Cl En
En ox	LiF	5	1,120	Pr En+Cl En
Nat En	LiF	5	1,120	Pr En
Cl En	LiF	5	1,120	Pr En+Cl En
Cl En	Li_2CO_3	7	1,140	Pr En
Cl En	NaF	7	1,140	Pr En+Cl En
En ox	LiF	7	1,150	Cl En
Cl En	LiF	7	1,150	Pr En+Cl En
Nat En	LiF	7	1,150	Pr En
Cl En	LiF	6	1,170	Cl En
Nat En	LiF	6	1,170	Cl En+Pr En
Nat En	None	9	1,175	Pr En
En ox	LiF	1	1,245	Pr En
Nat En	None	1	1,250	R En+Cl En
Nat. Anthophyllite	None	16 hours	1,250	Pr En
En gl	None	1	1,310	Pr En
En gl	LiF	1 hour	1,340	Pr En

* For purposes of compactness, the following abbreviations are used: R En = rhombic enstatite; Pr En = protoenstatite; Cl En = clinoenstatite; nat = naturally occurring; En gl = $MgSiO_3$ glass; En ox = a mixture of oxides having the composition $MgO \cdot SiO_2$.

The same thermal results may be fitted into an entirely different scheme of equilibrium relations:



It is seen that the only two stable phases are rhombic enstatite and protoenstatite; clinoenstatite is a metastable low-temperature phase occupying a position somewhat analogous to that of low cristobalite. In this scheme the presence of protoenstatite above 1,160° in some of the italicized runs is due to preservation of the stable high-temperature state by the quench. The clinoenstatite occurrences above and below 1,160° can be explained by assuming that in these runs the high-temperature state was not preserved by the quenching process or that the quenching process itself is responsible for the protoenstatite→clinoenstatite transition. The apparently erratic appearance of clinoenstatite does follow a more or less definite pattern. It may be noted that clinoenstatite becomes more common as the temperature of the run increases. Also that phase usually results from runs starting with the oxides or natural enstatite. Glass containing runs fired for relatively short lengths of time tends to preserve protoenstatite. It is reasonable to suppose that the different conditions of crystallization cited above have their effects on the size, perfection, habit, and surface properties of the crystals formed and that these properties may influence the preservation, or lack of it, of protoenstatite on rapid cooling. The term "rapid cooling" implies any rate of cooling which is fast enough to prevent reversion to rhombic enstatite.

The second equilibrium pattern is favored by certain experimental evidence and by relations between the hypothetical structures of protoenstatite and clinoenstatite. It has been observed by Büssem, Schusterius, and Stuckardt and by Clark that protoenstatite spontaneously transforms to clinoenstatite at room temperature in a few months; the transformation is attended by a disintegration of the polycrystalline mass. The same phenomenon has been observed in many runs which yielded clinoenstatite as a final product. This product commonly occurs as a loose powder or as a friable mass; runs resulting in rhombic or protoenstatite always occur as firmly coherent masses. This disintegration of clinoenstatite products is most marked in runs in which clinoenstatite was reheated to a temperature above 1,160°. A reasonable assumption is that the initial clinoenstatite inverted to protoenstatite on heating and then reverted to clinoenstatite on cooling; the second transformation causes the disintegration because there is insufficient time at high temperature to allow recrystallization of the final clinoenstatite into a coherent mass.

Further evidence in favor of the second pattern is given by the current work of W. R. Foster (written communication, November, 1950). Employing high-temperature X-ray powder techniques, Foster found that synthetic clinoenstatite inverted to protoenstatite at 1,350° C. in 1 hour. On the basis of this and other runs, he arrived independently at an equilibrium scheme almost identical with that of the writer.

A comparison of the structures shown in figure 1 certainly favors protoenstatite as the highest-temperature phase; its structure is the least sheared, and the volume of its unit cell is greatest relative to comparable units in the other two phases. In general, glasses tend to crystallize first in the highest-temperature form, which then transforms to the phase stable in the given region. The fact that $MgSiO_3$ glasses tend first to crystallize as protoenstatite provides some additional evidence in favor of the high-temperature stability of that phase.

In the section on structural relations, statements were made that atomic posi-

tions in the protoenstatite were between those in rhombic and clinoenstatite; i.e., during the process of conversion from rhombic to clinoenstatite or vice versa, the atoms pass through the positions they would occupy in protoenstatite. This is borne out by some of the results in table 2. It may be noted that many of the runs yielding clinoenstatite from rhombic enstatite and vice versa contain some protoenstatite. It was shown in the same section that many intermediate states between any two structures were theoretically possible. Thermal results indicate that none of these are stable in any temperature interval. Thus protoenstatite heated in the rhombic stability area gives a mixture of protoenstatite and rhombic enstatite, or, more likely, a series of intermediate states between those two structures. Nevertheless, at the same temperature, rhombic enstatite remains in that form; i.e., it does not transform to intermediate states. The same relations between proto- and clinoenstatite and between rhombic and clinoenstatite may be seen in the data of table 2.

In summary, the preponderance of evidence favors the existence of two stable phases of $MgSiO_3$, rhombic enstatite occurring up to 985° C. and protoenstatite occurring from 985° up to the point of dissociation into forsterite and a liquid. Clinoenstatite is a metastable low-temperature phase formed from protoenstatite by rapid cooling. The transformations from one phase to another are first-order transitions, although unstable intermediate states are possible. The transition rhombic enstatite $\rightleftharpoons$ protoenstatite is reconstructive, inasmuch as it is accelerated by a flux, whereas the transformation of protoenstatite to clinoenstatite is apparently displacive.

The Subsolidus System $MgSiO_3$-$CaMgSi_2O_6$

INTRODUCTION

The second part of this investigation will be devoted to an experimental determination of phase equilibria in the system $MgSiO_3$-$CaMgSi_2O_6$ below the liquidus region. With the exception of investigations of the polymorphism of $MgSiO_3$ and of the solid-solution relations just below the liquidus surfaces, the emphasis in this system in the past has been on phase equilibria in the presence of a melt.

Perhaps the first major experimental investigation of the system was that of Allen, White, Wright, and Larsen (1909). Two stable and three unstable crystalline forms of $MgSiO_3$ were postulated, none of which, according to these authors, was able to dissolve more than about 3 or 4 per cent of $CaMgSi_2O_6$. On the other hand, it was found that at about 1,375° C. the latter compound was capable of holding somewhat more than 60 per cent of its own weight of $MgSiO_3$ in solution. Higher percentages of $MgSiO_3$ resulted in inhomogeneous crystals.

Utilizing the newly developed quenching technique, Bowen demonstrated that the system must be considered as having three components because of the dissociation of $MgSiO_3$ to Mg_2SiO_4 and a liquid. The component $\alpha MgSiO_3$ was shown to be identical with forsterite, the true composition being Mg_2SiO_4. Bowen (1914) found that a complete solid-solution series existed between clinoenstatite and diopside; the inhomogeneities found by Allen, White, Wright, and Larsen (1929) for high percentages of $MgSiO_3$ consisted of tiny crystals of forsterite and SiO_2, not clinoenstatite. It

is of interest to note at this point that none of Bowen's listed runs in the solid-solution field was quenched at a temperature more than just a few degrees below the liquidus surface. Solubility relations for the other phases of enstatite were not considered in Bowen's paper.

A considerable amount of work has been done with natural rocks, with the purpose of determining the courses of crystallization of basaltic magmas. Part of this general problem has been the qualitative consideration of equilibria among the pyroxene phases. An excellent summary of investigations in this field has been given by Hess (1941). The results of these investigations and of his own work have been presented by Hess in a series of hypothetical phase diagrams whose similarity to the one to be presented later in this paper is striking.

ANALYSIS OF PHASES

Although various fluxes were used, it was found that most runs yielded crystals of such small size that optical analyses were impractical; only in parts of the clinopyroxene fields were crystals of suitable size obtained. The simplest method of analysis of phase mixtures and solid solutions was by means of X-ray powder photographs. There is little difficulty in determining the identity of the phases in a mixture of two or three components by this method, provided that 10 per cent or more of a given phase is present. The compositions of solid solutions in certain composition intervals were determined by the usual method of measuring shifts in spacings. Unfortunately, the unit-cell dimensions and consequently the interplanar spacings of diopside differ by only tenths or hundredths of an Angstrom unit from those of clinoenstatite. Therefore, most line shifts were of the order of tenths of a millimeter for a 10 per cent change in composition (on a powder film taken in a camera of 115-mm. diameter). However, one line of unknown index far in the back reflection area showed a shift of approximately 1.5 mm. per 10 per cent change in composition. This line appears on films covering the composition range En 50 per cent–Di 50 per cent to Di 100 per cent; for pure diopside its spacing is 0.976 Å. A line having shifts of the same magnitude appears on films of rhombic enstatite (d = 0.978 Å for 100 per cent En); thus the amount of diopside held in solid solution in that phase may be determined readily. In order to calibrate the line shifts, a series of standards starting with glass or oxide mixtures of known composition were run within the solid-solution fields. These runs were then mixed with a simple substance whose interplanar spacings were accurately known from the literature, and then powder photographs were taken of the mixtures. Since LiF of reagent grade had been used as a flux and because its spacing pattern is known,[6] it served very well as a standard. Standard curves for the analysis of solid solutions are given in figures 2 and 3. The ordinate, $\Delta 4\theta$, is four times the difference between the glancing angle of the standard LiF line (d = 1.004 Å) and that of the diagnostic line. Owing to the diffuseness of lines in the back reflection area, it is estimated that a composition determination based on the standard curves involves an uncertainty of about ± 3 per cent.

At high temperatures crystals suitable for analysis by the usual optical methods were obtainable. Analyses were carried

[6] The spacings for LiF were taken from the *Alphabetical Index of X-ray Diffraction Patterns*, published by the American Society for Testing Materials, 1945.

out by a determination of the index of refraction, γ, and of the extinction angle, $Z\wedge c$; the results were compared to the standard curves given in Winchell (1931). The index of refraction was measured by a single variation technique employing a 5-axis universal stage and sodium D illumination. Crystals were immersed in oil whose index was varied by changing the temperature. When a perfect match had been obtained, the index of the oil at the given temperature was directly read from an Abbe refractometer. The above method is an adaptation of the double-variation technique described by Emmons (1943). The extinction angle of the oriented grain was measured directly. The reliability of analyses performed in the manner described above is within ± 4 per cent.

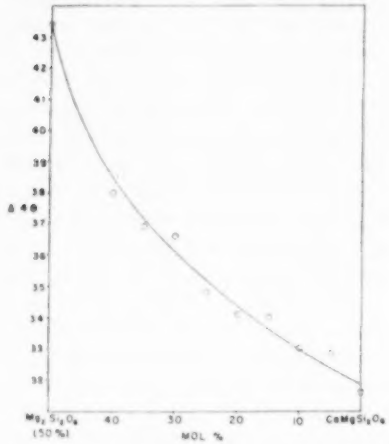


FIG. 2.—Curve giving the difference in glancing angles between the diagnostic reflection of diopside solid solution and a neighboring LiF reflection as a function of composition.

PHASE EQUILIBRIA

EXPERIMENTAL PROCEDURE

The basic starting materials which were subjected to thermal treatment were glasses and stoichiometric mixtures of oxides, all having known compositions relative to the percentages $Mg_2Si_2O_6$ and $CaMgSi_2O_6$ "molecules." The runs were air-quenched, and the results were analyzed by the methods described above. Wherever possible, attempts were made to establish reversibility; i.e., starting materials consisting of high-temperature phases synthesized from oxides or glasses were heated in lower-temperature phase fields. It was found that these reactions proceeded at only a few points on the diagram. Two different methods were used to locate boundary curves. The first and most accurate method was useful in determining solid-solution boundaries. The technique consisted of making a run in an immiscibility region such that the composition of the starting material was within 10 or 20 per cent of the curve to be located. The resulting material was then analyzed for the composition of one of the two solid-solution phases present. This value yielded a point on the boundary curve. Runs were made fairly close to boundary curves, so that one phase would predominate; this insured that diagnostic X-ray powder lines of one phase would not be obscured by those of the other phase.

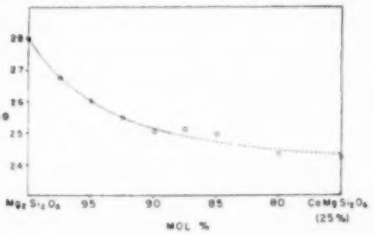


FIG. 3.—Curve giving the difference in glancing angles between the diagnostic reflection of rhombic enstatite and a neighboring LiF reflection as a function of composition. The dashed portion of the curve was given by unstably crystallized rhombic enstatite solid solutions.

The second method consisted of limiting the stability fields by the first appearances of new phases or by changes in the coherency of the final crystalline mass. Runs were made at constant composition and at increasing or decreasing temperatures until examination under the microscope or the appearance of new lines on a powder photograph indicated the presence of a new phase. This procedure involves considerable doubt, particularly in the placement of protoenstatite field boundaries, because of the difficulty in preserving that phase with a quench. It was generally assumed, therefore, that, if a run contained clinoenstatite and the charge had decrepitated, the point so determined was in the protoenstatite stability region. This method of placing phase boundaries involves considerable uncertainty; therefore, they will be represented by dashed lines. It is clear that, if a run contains clinoenstatite and another phase, the second phase may act as a bonding material. Thus the crystalline mass may be coherent, even though the clinoenstatite had formed during the quench.

EXPERIMENTAL RESULTS

The results of thermal experiments are presented in table 3 and in figure 4. It will be noted from table 3 that the determination of the stability of protoenstatite solid solutions is based largely on the physical condition of runs containing clinoenstatite after quenching. In order to locate the curve *FG*, the assumption has been made that during the quenching process, with the attendant transformation of protoenstatite to clinoenstatite, no change in the solubility relations occurs. The analyses were necessarily carried out on the clinoenstatite which is actually found in the cooled runs.

DISCUSSION

The phase equilibria in the enstatite-diopside system may perhaps be best discussed by analyzing the course of phase changes in figure 4 for two compositions. Consider a composition 60 per cent En, 40 per cent Di. Above point *X* a homogeneous solid solution exists. As the temperature is lowered below point *X*, exsolution occurs, so that clinoenstatite solid solution coexists with a diopside solid solution. The compositions of these two phases are given by the two sides of the immiscibility curve. At point *Y*, the clinoenstatite solid solution is converted to a protoenstatite solid solution. When the temperature reaches the value at *Z*, the protoenstatite is transformed to rhombic enstatite solid solution. Below *Z* the latter phase coexists with diopside solid solution. The compositions of these solid solutions are given by curves *AB* and *DE*.

Above point *L*, a composition corresponding to 95 per cent En and 5 per cent Di exists as a homogeneous clinoenstatite solid solution. At *L* a protoenstatite solid solution separates out and coexists with the clinoenstatite. When point *M* is reached, all the material is converted to protoenstatite solid solution, which is stable until the temperature drops to *N*. At this point rhombic enstatite solid solution first forms and then exists stably with protoenstatite down to point *O*. From *O* to *P*, rhombic enstatite is the only stable phase. Finally, at *P* diopside solid solution exsolves and coexists with rhombic enstatite.

Several interesting relationships are shown in the phase diagram. The slopes of the protenstatite and rhombic enstatite solid-solution curves at the extreme left of the figure indicate that the large Ca atom is less soluble in the high-

TABLE 3

THERMAL DATA FOR THE SUBSOLIDUS SYSTEM $CaMgSi_2O_6$-$Mg_2Si_2O_6$

INITIAL MATERIAL*	$Mg_2Si_2O_6$ (MOL PER CENT)	TIME (DAYS)	TEMP. (° C.)	FINAL PHASES*
	Curve *ABC*			
Glass	20	2	725	Di ss(3% En)+R En ss
Glass	20	2	785	Di ss(5% En)+R En ss
Glass	30	2	830	Di ss(9% En)+R En ss
Glass	30	1	890	Di ss(9% En)+R En ss
Glass	30	1	940	Di ss(12% En)+R En ss
Oxides	60	1	985	Di ss(15% En)+R En ss
Oxides	40	1	1,005	Di ss(18% En)+R En ss
Oxides	50	1	1,060	Di ss(20% En)+R En ss
Glass	40	1	1,090	Di ss(24% En)+R En ss
Oxides	70	1	1,125	Di ss(26% En)+Cl En ss
Di ss	50	3	1,160	Di ss(28% En)+Cl En ss
Oxides	40	1	1,190	Di ss(29% En)+Cl En ss
Oxides	40	1	1,205	Di ss(32% En)+Cl En ss
Oxides	50	1	1,230	Di ss(33% En)+Cl En ss
Oxides	45	1	1,255	Di ss(36% En)+Cl En ss
Glass	60	1	1,300	Di ss(39% En)+Cl En ss
Glass	60	1	1,320	Di ss(43% En)+Cl En ss
Oxides	60	1	1,350	Di ss(50% En)+Cl En ss(68%)
	Curve *DE*			
Glass	92.5	5	750	R En ss(95% En)+Di ss
Glass	82.5	2	805	R En ss(98% En)+Di ss
Oxides	80	3	860	R En ss(97% En)+Di ss
Oxides	80	2	900	R En ss(97% En)+Di ss
Glass	82.5	2	960	R En ss(96% En)+Di ss
Glass	80	1	1,010	R En ss(95% En)+Di ss
Glass	87.5	8	1,050	R En ss(93% En)+Di ss
Glass	80	5	1,075	R En ss(91% En)+Di ss
	Curve *FGH*			
Glass	80	12	1,140	Cl En ss(88% En)+Di ss(D)
Glass	60	12	1,175	Cl En ss(88% En)+Di ss
Glass	70	5	1,200	Cl En ss(88% En)+Di ss
Glass	60	1	1,235	Cl En ss(83% En)+Di ss
Glass	60	4	1,250	Cl En ss(83% En)+Di ss
Glass	60	1	1,285	Cl En ss(83% En)+Di ss
Glass	60	3	1,295	Cl En ss(78% En)+Di ss
Glass	60	1	1,320	Cl En ss(78% En)+Di ss

* Abbreviations are the same as in table 2, with the following additional ones: Di = diopside ($CaMgSi_2O_6$); ss = solid solutions; (D) indicates that the charge had decrepitated.

All solid solutions containing less than 60 mol per cent $Mg_2Si_2O_6$ will be called "diopside solid solutions" (60 per cent En represents the maximum of the immiscibility curve).

Solid solutions containing more than 60 per cent En will be designated as "protoenstatite solid solutions" or "clinoenstatite solid solutions."

TABLE 3—*Continued*

Initial Material*	$Mg_2Si_2O_6$ (Mol Per Cent)	Time (Days)	Temp. (° C.)	Final Phases*
			Line *FEB*	
Glass	87.5	12	1,135	Cl En ss+Di ss+R En ss
Glass→Cl En	85	7	1,130	Cl En ss+Pr En ss+Di ss
Glass	80	12	1,140	Cl En ss+Di ss
Oxides	70	1	1,125	Cl En ss+Di ss
Glass	60	12	1,140	Cl En ss+Di ss
Oxides	60	1	1,125	Cl En ss+Di ss
Oxides	60	1	1,115	Cl En ss+Di ss
Oxides	50	1	1,135	Cl En ss+Di ss
Oxides→R En	80	5	1,075	R En ss+Di ss+Cl En ss
Glass	80	5	1,075	R En ss+Di ss
Oxides→R En	70	5	1,075	R En ss+Di ss+tr Cl En ss
Oxides	60	5	1,075	R En ss+Di ss
Oxides	60	1	1,050	R En ss+Di ss
Oxides	50	1	1,085	R En ss+Di ss
			Line *ST*	
Oxides	85	1	1,160	Cl En+Di ss (D)
Oxides	80	1	1,160	Cl En+Di ss (D)
Oxides	70	1	1,160	Cl En+Di ss (D)
Glass→R En	80	1	1,205	Cl En+Di ss (D)
Oxides	70	1	1,205	Cl En+Di ss (D)
Glass→R En	80	1	1,250	Cl En+Di ss partial (D)
Oxides	70	1	1,250	Cl En+Di ss coherent
			Region between 90% En and 100% En and from 985° C. Upward	
Oxides	97.5	5	1,010	Pr En+R En
Glass→Pr En	97.5	7	1,020	Pr En+R En
Glass	97.5	5	1,025	Pr En+R En
Glass→Cl En	97.5	10	1,080	Cl En+Pr En
Glass	97.5	8	1,170	Pr En
Glass→R En	95	12	1,035	R En+tr Cl En
Glass→R En	95	12	1,055	Cl En (D)
Glass	95	2	1,105	R En+Pr En
Glass	95	4	1,150	Pr En+Cl En (D)
Glass	95	5	1,190	Cl En (D)
Glass→Cl En	95	1	1,275	Pr En+Cl En
Glass→Cl En	95	1	1,305	Cl En (D)
Glass→R En	92.5	1	1,305	Cl En
Glass	92.5	12	1,055	R En
Glass	92.5	5	1,080	R En
Glass	92.5	6	1,115	Cl En (D)
Glass→Cl En	90	5	1,095	Cl En+Pr En (D)

temperature form than in the low-temperature phase. Although data are lacking because of the inadequacy of the quenching technique used, a similar set of curves having slopes in the opposite direction probably bound the protoenstatite and clinoenstatite solid-solution fields. It is thus evident that Ca is also less soluble in protoenstatite than in clinoenstatite. Apparently, if sufficient Ca is available, clinoenstatite becomes a truly stable high-temperature phase. The entrance of Ca into the clinoenstatite structure increases the angle β, eventually causing the interchange of Z co-ordinates of the two central chains shown in figure 1, *B*. The structure then becomes analogous to that of diopside, differing only in the value for angle β. It is possible that this reversal in the Z co-ordinates of the central chains may have much to do with the stabilization of clinoenstatite solid solution.

GEOLOGIC APPLICATION

The enstatite-diopside system considered in this investigation includes two components of the important four-component geologic system: diopside-hedenbergite-enstatite-ferrosilite. Because Fe readily substitutes for Mg in enstatite and diopside, it is likely that replacement of up to 50 per cent of the Mg by Fe in those two phases would have little

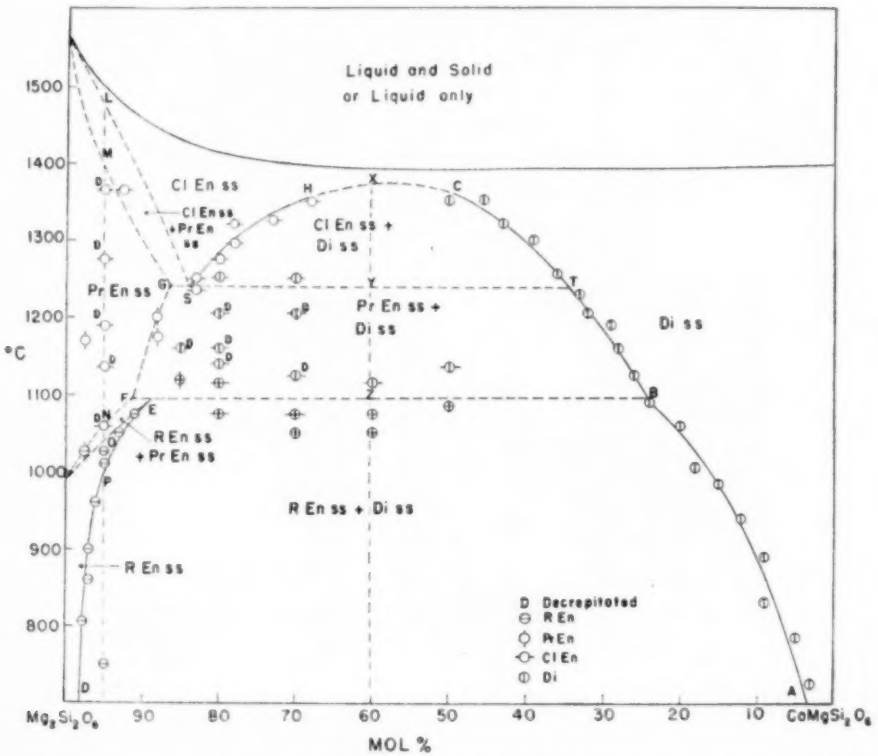


FIG. 4.—Phase diagram for the subsolidus system $CaMgSi_2O_6$-$Mg_2Si_2O_6$. Dashed lines indicate uncertainty in the location of the curves in question. The liquidus curve was adapted from Bowen (1914).

effect other than to shift slightly the phase diagram of figure 4 to lower temperatures. Therefore, the diagram may possibly be useful, in so far as curves *AB* and *DE* are concerned, to indicate a maximum-temperature value for the last equilibrium state of rocks containing enstatite and diopside solid-solution phases. With the same limitations, the rhombic enstatite-protoenstatite inversion point may also serve as a temperature indicator. Of great interest also is the fact that that inversion may provide information regarding the rate of cooling of a rock formed above the inversion temperature. One would expect that, in a rock that had cooled slowly, the protoenstatite would have transformed to rhombic enstatite. If, however, the cooling had been rapid, the protoenstatite would have transformed into the metastable clinoenstatite. This conclusion is borne out by the fact that reported natural occurrences of clinoenstatite have been from meteorites and lavas only; to the writer's knowledge, no occurrences have been listed from hornfels zones or from intrusive igneous bodies, where temperatures may have been above the transition point.

ACKNOWLEDGMENTS.—The writer wishes to express his indebtedness to the faculty of the Department of Geology of the University of Chicago for the laboratory facilities and financial assistance which made this research possible. He is particularly grateful to Dr. Julian R. Goldsmith for his guidance and for his invaluable advice on matters of equipment and technique. Much appreciation is felt for the patient counsel on the use of X-ray diffraction methods given by Dr. Fritz Laves, Dr. D. Jerome Fisher, and Dr. Alfred J. Frueh. Dr. Hans Ramberg suggested the subject of this investigation to the writer.

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SURFACE AREA OF DEEP-SEA SEDIMENTS[1]

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ABSTRACT

Absolute surface areas of a set of seventy-five representative deep-sea core samples have been measured by the gas-adsorption method. The values range from 2.5 m^2/gm for fine red silt to 48.0 m^2/gm for some continental clay but are characteristic of a specific sediment type. In the most common sediment, a pink to gray foraminiferal clay, the surface areas vary from about 7 to about 30 m^2/gm, depending largely on the concentration of the Foraminifera shells.

Pertinent aspects of the theory and the technique of this method of surface-area measurement are presented.

INTRODUCTION

One of the potentially valuable methods of determining the age of layers of deep-sea sediments is the ionium, or radioactive inequilibrium, method, suggested by William Urry (Piggot and Urry, 1944). This method is based on the assumption that a constant excess of ionium (thorium 230) over the amount in equilibrium with uranium is incorporated into a sediment of a specified mineralogical type. Thus a plot of ionium content against depth should show the ionium decay curve (half-life, 83,000 years) from which age at a given depth can be calculated.

Direct application of this method to a variety of north Atlantic cores is in progress; however, it is essential that the mechanism of the ionium incorporation be understood if the limitations of the method are to be defined. For a constant excess of ionium to be present in a sediment, a constant process must be operating in a homogeneous environment. It is reasonable that relative concentrations of ionium and uranium (environment) have remained constant through several hundred thousand years, but it is less probable that the physical and chemical characteristics of the sediment have remained invariant. The method obviously will have validity only for a plot of ionium against depth for a given sediment type. Hence the physical and chemical characteristics which may affect the process must be determined. If the mechanism which produces the excess ionium is adsorption, it is reasonable to assume that the quantity adsorbed might be directly related to absolute surface area. If the type of site required to adsorb an ionium ion can be defined by other experiments, the number of sites available per gram of sediment can be calculated from the surface area.

Further, the surface area of the deep-sea sediments may be of considerable value in other types of basic studies of the geology of the ocean bottom. Therefore, it was decided to measure the surface areas of a representative suite of specimens from various deep-sea cores. The results will be interpreted for their implication to the ionium age method in a later paper when the radioactivity data are completed. This report is primarily concerned with the theory and measurement of surface area.

THEORY

A brief review of the terms and concept of adsorption may assist in understanding the discussion which follows. If

[1] Lamont Geological Observatory Contribution 14. Manuscript received February 10, 1951.

a gas is incorporated in the inside of a solid, it may either dissolve, thus forming a solid solution, or it may react to form a compound. In either case it is called "adsorption." Likewise, a gas remaining on the surface of a solid may be attracted slightly in a manner similar to condensation, or it may react chemically with the surface with a much higher bonding energy. The former is called "physical" or "Van der Waals adsorption," while the latter is termed "chemical adsorption" or "chemisorption." The surface-area measurement is made by the use of physical adsorption because such adsorption is not selective as to sites above a certain minimum pressure.

When a gas (adsorbate) is admitted to an evacuated solid (adsorbent), some of the gas is adsorbed on the surface of the solid, and the rest remains in the gas phase. The relative amounts are determined by the temperature, pressure, and nature of the gas and the solid. In physical adsorption this distribution takes place quite rapidly—on the order of 15–30 seconds. This type of adsorption will take place between any surface and any gas if the temperature is low enough. A plot of the amount of gas adsorbed against the pressure for a given gas and unit weight of solid at a fixed temperature is called an "adsorption isotherm."

The formulation of the Brunauer, Emmett, and Teller (1938) theory of multimolecular adsorption and the previous experimental work and observations on gas adsorption by Brunauer and Emmett (1935, p. 1754; 1937, p. 2682; Emmett and Brunauer, 1937, p. 1553) presented chemists with a method of absolute surface-area determination of extraordinary versatility. In principle, the method depends on the measurement of the volume of a certain gas required to cover a surface to a depth of a monomolecular layer. Then, knowing the area covered per molecule, it is possible to calculate absolute surface area. The method is simpler to apply and far more accurate than other methods of surface-area determination for fine-grained material. Actually, it is universal in application in regard to both the extent of the surface and the chemical nature of the surface. Absolute values of surface area are probably correct to 20 per cent, whereas relative values obtained with different adsorbent gases at different temperatures agree to within 10 per cent. For a given experimental arrangement operating with a single gas at a fixed temperature, the reproducibility is 1–2 per cent. In order to obtain the best value for the actual surface of an adsorbent, the smallest molecules should be used (hydrogen and helium); but, because it is necessary to work below the boiling point of the gas, one usually uses argon or nitrogen at liquid-air temperatures. The difference in surface area for most materials is certainly not significant. The derivation of the Brunauer, Emmett, and Teller theory and a brief discussion of suggested modifications are given in the appendix.

EXPERIMENTAL PROCEDURE

The system (fig. 1) consists of an oil (*1*) and mercury (*2*) vacuum pump in series, followed by a dry-ice trap (*3*), a McLeod gauge (*7*), an argon manometer (*5*), adsorption bulb (*4*), and gas burette (*6*). Purified tank helium and spectroscopically pure argon are stored in the bulbs above the manifold.

The samples of deep-sea sediments were taken from cores obtained by Professor M. Ewing on recent Atlantic expeditions. Preliminary tests showed a difference in surface area of less than 10 per cent in a sample that was lightly

broken up to fit in the adsorption bulb and the same sample after it had been ground to fine powder with a mortar and pestle. The procedure of grinding the sample to pass a 50-mesh screen was adopted as standard, since this size proved most satisfactory for alpha counting on the same samples.

The sample, weighing about 1 gm., was first heated for 2 hours at 150° C. under vacuum in the adsorption bulb (*4*). Heating for 12 hours at 280° C. produced no change in the surface area. Therefore, 2 hours at the lower temperature was adopted as sufficient to remove all adsorbed water as well as volatile organic impurities from the sample. The ground joint of the adsorption bulb was

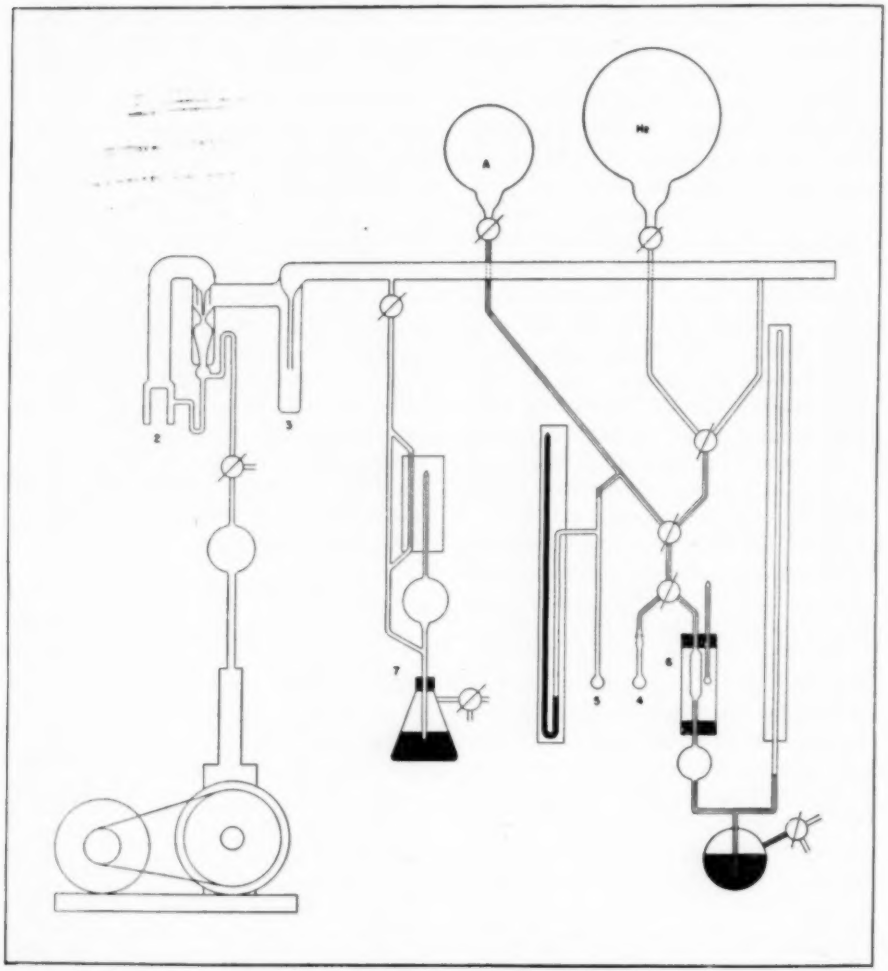

FIG. 1.—Surface-area measurement apparatus

cooled by a stream of air during the heating, to prevent any leakage. After the sample had cooled to room temperature, the entire system was pumped to better than 10^{-5} mm. Next the "dead space" of *4* is determined by expanding a known amount of helium from the gas burette (*6*) into the adsorption bulb (*4*), with the adsorption bulb surrounded by liquid N_2. This is repeated three times. The helium is then pumped out of the system, and argon is admitted to *5* in sufficient quantity to produce a liquid phase with the liquid nitrogen (*4* and *5*). The vapor pressure of the argon at this temperature, p_0, is read on the manometer (*5*). The adsorption isotherm of argon on the sediment is then determined by expanding successive quantities of argon from *6* into *4*. The following relation indicates how the amount adsorbed is determined:

$$P_{\substack{\text{initial}\\(\text{burette})}} V_{\text{burette}} - P_{\text{final}} V_{\substack{\text{dead space}\\+\text{burette}}} = P V_{\text{adsorbed}} .$$

The process is continued until the final pressure after adsorption is such that p/p_0 is greater than 0.30. At higher values the simple BET plot is not linear. The volume adsorbed is thus calculated for each final pressure and converted to S.T.P. Now there are sufficient data in the correct form to obtain the values for $p/v(p_0-p)$ and p/p_0 so that the BET plot can be made. The reciprocal of the slope is the volume of gas required for a monolayer. The cross-sectional area for the argon atom is assumed to be 12.9 sq. Å, after Armbruster and Austin (1939, p. 1117). Therefore,

$$\frac{V_m \times \text{area/mole } (m^2)}{\text{cc/mole at S.T.P.} \times \text{sample weight}} = \text{Surface area } (m^2/gm) .$$

Figure 2 shows the BET plot for three representative core samples. They were chosen to illustrate the considerable range in surface-area values. They also show the variation in the precision. The curve for A152-137-523 is typical of a careful measurement without experimental difficulties. This shows the remarkable linearity and the excellent precision obtainable with the method. On the other hand, the curve for C87-145 is about as poor in precision as is tolerable. Even with this spread of points, however, the surface area will be correct within 5 per cent.

TABLE 1

SURFACE AREAS OF VARIOUS SUBSTANCES

Adsorbent	Surface Area (M^2/Gm)
Mica (0.1-mm. sheets)	0.0036 (approx. geom. area)
Fe_3O_4 catalyst (unreduced)	0.02
$CuSO_4 \cdot 5H_2O$ (40–100 mesh)	0.16
KCl (finer than 200 mesh)	0.24
Pumice	0.38
Cement	1.08
$BaSO_4$ (pptd.)	4.30
Montmorillonite and kaolinite	15.5 (15.3)
Cecil soil 9418	32.3
Halloysite	43.2
Illite	97.1
Silica gel	584
Darco G	2,123

RESULTS

The BET surface areas of about 80 samples taken from fourteen different cores have been measured. These samples were selected to yield information on as wide a range of sediment types as possible. In core A-152-118 a fairly detailed analysis throughout the length of the core was carried out. The results are summarized in table 2.

In order to compare the surface area of deep-sea sediments with that of other common materials, table 1 has been compiled. These data were obtained by other workers using the BET technique (Bru-

nauer, 1943; Nelson and Hendricks, 1943). Note the range in surface area from a geometrical example like mica, through ground cement, montmorillonite, illite, silica gel, and Darco G. In the case of Darco G, almost every other carbon atom is on the surface.

In table 2 the columns give, in order, the core number, its location, the depth of water, the point at which it was sampled, expressed in centimeters from the top, the measured surface area, and a brief description of the core at that point. It is evident that the surface area of the deep-sea sediments may vary by a factor of 10. Variations up to 30 per cent occur even in such homogeneous cores as C-8-7. In this core there is an unconformity at 335 cm. The pink clay below 335 cm. is somewhat finer than the green above and has cold-water fauna. The sample taken at 343 cm. contained suf-

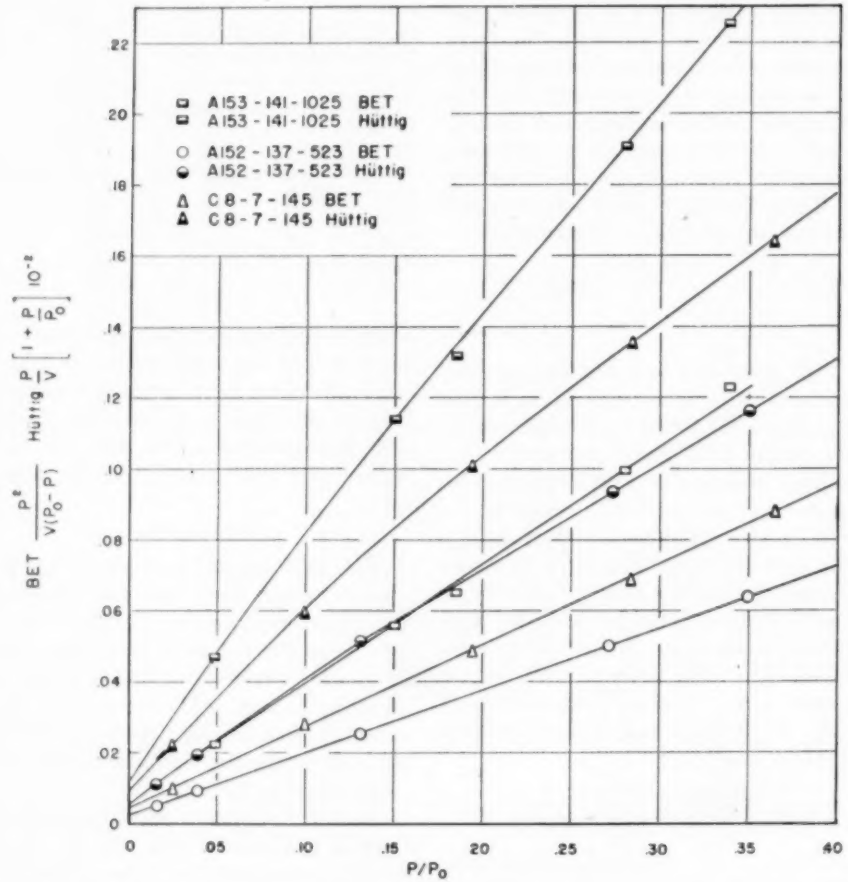

FIG. 2.—Adsorption isotherm data for three representative core samples plotted according to the BET and Hüttig equations.

TABLE 2

SPECIFIC SURFACE AREAS OF DEEP-SEA SEDIMENTS

Core No.	Depth (Cm.)	Location	Water Depth (M.)	Surface Area (M^2/Gm)	Description
C8-7	26	35°56′ N., 74°41′ W.	1,370	10.5±0.4	Typical green clay
	53			12.7±0.5	
	99			12.1±0.5	
	130			11.7±0.5	
	145			12.5±0.5	Continental slope
	188			10.3	
	227			11.1±0.4	
	330			14.0±0.5	
		(Unconformity)			
	340			15.5±0.5	Pink clay similar above
	343			10.7±0.4	Pink clay with some sand
	361			17.3±0.5	Pink clay
	487			12.6±0.5	
A152-84	100	44°21′ N., 30°16′ W.	2,750	23.6±0.7	Calcareous clay, detritus,
	280			18.7±0.6	cold-water fauna (Wisc.?)
	311			6.2±0.3	
	411			9.2±0.3	Globigerina ooze, warm wa-
	450			10.0±0.2	ter (Sangamon?)
A152-88	94	39°5½′ N., 26°25′ W.	2,380	7.1±0.3	Volcanic ash layer
	262			11.2±0.4	$CaCO_3$ Foraminifera and coccoliths
	600			7.8±0.3	Mostly Foraminifera
A152-127	49	34°56′ N., 46°19′ W.	4,630	29.8±0.8	Pink foraminiferal clay
	270			30.2±0.8	Gray foraminiferal clay
	649			36.7±0.8	Red clay
A152-135	348	36°31′ N., 67°31′ W.	4,850	16.5±0.5	Silty gray clay
	710			20.8±0.6	Gray clay
A152-137	18	38°5′ N., 69°35′ W.	3,800	10.2±0.4	Gray foraminiferal clay
	523			25.6±0.7	Red clay
	630			24.2±0.6	Red clay breccia (slump?)
A153-141	745	33°26½′ N., 53°48′ W.	5,350	2.5±0.1	Fine red sand with minor
	1025			5.4±0.1	clay
A156-2	70	29°12½′ N., 76°49′ W.	2,140	38.6±0.9	Fine Eocene coccolith mate-
	172			36.6±0.9	rial ($CaCO_3$)
A156-12	30	38°23′ N., 70°57′ W.	3,470	4.3±0.2	Sandy layer
	68			17.2±0.5	Pink foraminiferal clay
	308			48.0±0.9	Clay ball in gravel (trans.?)
C10-10	182	32°19′.4 N., 64°35′.6 W.	1,005	3.2±0.1	Coarse $CaCO_3$ (*Amphistegina*)
C10-5	115	33°37′.4 N., 62°29′.6 W.	2,670	25.7±0.5	Bentonite, altered igneous rock
	211			12.5±0.4	Globigerina with coccoliths
	430			5.0±0.2	Coarse Foraminifera

TABLE 2—*Continued*

Core No.	Depth (Cm.)	Location	Water Depth (M.)	Surface Area (M^2/Gm)	Description
A157-5.....	10	48°35′ N., 36°51′ W.	4,500	18.8	
	23			27.8	Foraminiferal gray clay
	43			22.5	
	63			24.5	
	84			28.8	Light-tan clay and sand grains
	112			32.7	Bluish clay with minor Foraminifera
	155			18.3	Light-tan clay and sandy silt
	186			14.2	Gray clay and Foraminifera
	222			31.1	Glacial marine clay and silt
	248			17.8	Gray foraminiferal clay
A152-118....	11	35°7′ N., 44°40′ W.	4,340	13.4	Light-pink globigerina ooze
	25			17.9	Increase in clay content
	30			34.1	
	40			29.7	
	52			27.1	Calcareous red clay
	75			32.5	
	110			21.9	
	128			25.2	
	147			23.3	Gray clayey globigerina ooze
	150			21.6	
	181			21.7	
	200			23.7	
	210			24.8	
	234			25.0	
	263			25.3	
	285			22.1	
	305			18.0	Gray clayey ooze
	360			15.8	
	380			18.8	Pink clayey ooze (Foraminifera)
	400			30.1	
	407			24.2	Red calcareous clay
	410			28.4	
	420			26.2	
	442			27.6	Gray calcareous clay
	539			24.4	

ficient sand to decrease the surface area relative to the purer pink clay.

Core A152-84 illustrates the sharp contrast in surface areas of a calcareous clay with detrital material and cold-water fauna and a globigerina ooze made up of fairly large Foraminifera shells. These shells have essentially no internal surfaces and hence are as effective as rounded sand grains in lowering the surface area of the aggregate.

In core A152-88, a volcanic-ash layer at 94 cm. is clearly visible without the aid of a microscope. The surface area is comparatively low, for, although the surface area of fine volcanic ash must be

greater than that of pumice, it is glassy and probably has few internal surfaces. At 262 cm., calcium carbonate coccoliths surrounding large Foraminifera make up about 85 per cent of the material. Although the coccoliths have little internal surface, they are very fine particles. However, the forams cause a noticeable reduction in the surface area from that of a pure coccolith ooze.

Core A152-127 is very homogeneous and contains the finest clay observed in these cores. The surface area of 36.7 m^2/gm is correspondingly high.

Core A152-135 is a core taken at the foot of the Caryn sea mount beyond the continental slope. At 348 cm., it is a silty gray clay with about 20 per cent sand. At 710 cm., however, there is very little sand and not enough Foraminifera to lower the surface area appreciably.

Core A152-137 shows the effect of Foraminifera on the color and surface area of the sediment. In general, as the concentration of forams increases, the color becomes lighter and the surface area decreases. There is a sharp break in lithology at 50 cm. The deeper-water red clay appears always to have a higher surface area than the green or even the gray clay.

Core A153-141 is of fine deep-sea sands. The surface area at 745 cm., where the amount of clay is very small, is more than doubled at 1,025 cm. by the presence of about 10 per cent clay. (The term "clay" in this discussion refers to texture, not mineral composition. It appears that most of the "clay" of the deep-sea cores is rock flour. Kaolinite is rare, whereas illite and montmorillonite are present in some samples in minor amounts.)

Core A156-12, from Hudson Canyon, is heterogeneous. At 30 cm. there is a sandy layer, at 68 cm. a typical pink clay with some Foraminifera, and at 308 cm. a clay ball was found in a gravel layer. This plastic clay is probably continental kaolinite or montmorillonite.

Core C10-10 shows the low surface area of a chalky layer composed of coarse calcium carbonate *Amphistegina* shells mixed with volcanic material.

Core C10-5 has a texture and composition at 430 cm. similar to those of C10-10, but at 211 cm. the fine-grained coccolithic foraminiferal carbonate sediment is present. Near 100 cm. the core is composed largely of bentonitic material.

Core A156-2 is of considerable interest, in that it is composed of very fine Eocene coccolithic material. This material gave the highest surface area observed in this study.

A rather complete surface-area check was made of core A152–118. The surface areas can be understood when the effect of sediment type, coccoliths, and Foraminifera is considered. At the top the core is about 60 per cent calcium carbonate, a light-pink globigerina ooze with Foraminifera not abundant. The surface area is less than that for the pure clay because the clay is diluted with Foraminifera. At approximately 30-cm. depth the clay content of the sediment increases, and it becomes a darker red (14B7 Ridgeway); the surface area jumps to 34.1 m^2/gm. From 50 to 80 cm. the color becomes gradually lighter and increases in shell content. There is a marked change at 80 cm. The color at 108 cm. is 13A3 and the surface area at 110 cm. is 21.9 m^2/gm. From here down to 300 cm. the core is quite uniform, with a slight increment in clay content bringing the color to 13B5 at about 225 cm. and giving a surface area of 25 m^2/gm at 234 cm. From 300 to 380 cm. the sediment is a deep-water red clay with very few forams. The remaining samples were of a fairly

homogeneous red clay with a few fragmental forams.

Core A157-5 was also analyzed in some detail. The upper 20 cm. consists of a foraminiferal gray clay with a surface of 18.8 m^2/gm. Below 20 cm. the foram content drops off markedly, but the clay type appears to remain the same. From 75 to 225 cm. the material was inhomogeneous glacial marine sediment. The surface area depended in these samples on the percentage of light-tan (continental?) clay. At 248 cm. a gray foraminiferal clay was sampled and yielded a typical value for surface area for this type.

DISCUSSION

These data suggest the general pattern of surface-area distribution in deep-sea sediment types. Although the areas measured range from 2.5 to 40 m^2/gm and hence are significantly different, nevertheless they are more uniform than might have been expected. The relatively narrow range compared to the range of materials shown in table 1 is due to the sorting and sizing that occur in the long transportation process to the open ocean.

From the available data one may generalize the surface areas of certain sediment types. The surface area of fine sand or coarse forams (with minor clay) range from 1 to 5 m^2/gm. For the continental slope green clay, the values are from 10 to 14 m^2/gm. Sediment made up largely of small forams or of volcanic ash ranges from 5 to 10 m^2/gm. Typical globigerina ooze of foraminiferal gray clay ranges from 10 to 20 m^2/gm, depending on the foram content. The pure gray or light-pink deep-sea clay appears to have a surface area of about 30 ± 5 m^2/gm, decreasing with foram content and increasing with ferric oxide content. Fine coccolithic material may reach from 35 to 40 m^2/gm in surface area if pure. The percentage of this material in the ordinary globigerina ooze affects the surface area considerably.

As would be expected, the glacial marine deposits are inhomogeneous and show a wide variation in surface area, depending on the selection of the material. It appears that the plastic clay associated with these sediments is continental, with a surface area of about 40–50 m^2/gm.

The range of surface area shown by these materials is such that they could be described as moderately good adsorbents. It is to be expected from the polar nature of the minerals comprising the sediments that the entire surface would be covered by a layer of adsorbed ions. The particular concentration of a particular ion would be determined by the energy of adsorption and the relative concentration of that ion in sea water. But, as no satisfactory adsorption data are available in the low-concentration range of the radio-elements in sea water, it is not possible to evaluate the importance of surface area in the precipitation of these ions. If an equilibrium process exists, the surface area may be an important variable. However, if it is a rate process and there is more than enough surface on any of these materials to adsorb most of the radio-elements, then the extent of surface area has little significance. Experiments are now being conducted in this laboratory to study the adsorption of the radio-elements on deep-sea sediments.

CONCLUSION

The surface areas of a representative set of deep-sea sediment types have been determined to within an accuracy of 2 per cent. The values range from 2.5 to 40 m^2/gm but are characteristic of a

specific sediment type. This information, when coupled with adsorption data, may permit a definition of the mechanism by which ionium is removed from sea water.

ACKNOWLEDGMENTS.—The authors wish to acknowledge the helpful discussions with Mr. David Ericson on the material of this paper. Mr. Ericson also provided the lithologic description of the cores. The research reported in this document has been made possible through support and sponsorship extended by the U.S. Navy, Office of Naval Research, under Contract N6-onr-271, Task Order 18.

APPENDIX

Numerous minor modifications have been proposed for the BET theory (Brunauer, Deming, Deming, and Teller, 1940; Pickett, 1945; Andersen, 1946; Ross, 1948), but the absolute surface area is not affected by more than the 20 per cent originally suggested as the limit of absolute error by the authors of the theory. A recent paper by Halsey (1948) develops a new theory of physical adsorption on the basis of the nonuniformity of real surfaces. Halsey objects to the untenable hypothesis in the BET theory that an isolated adsorbed molecule can adsorb a second molecule on top, yielding the full energy of liquefaction, and that, in turn, the second molecule can adsorb a third, and so on. Halsey's reasoning would indicate that, below a p/p_0 of $\frac{1}{3}$, only a monolayer exists. However, Halsey admits that the surface-area determinations remain completely satisfactory. This is due in part to the fact that for ionic surfaces using a nonpolar gas as adsorbent, the heat of adsorption of the first layer is considerably larger than for subsequent layers.

The isotherm equation must now be derived. The basic assumption underlying the theory of multimolecular adsorption is that the forces are Van der Waals in character, e.g., those acting in condensation to the liquid phase.

Let $S_0, S_1, S_2, \ldots, S_i$ represent the surface area that is covered by $0, 1, 2, \ldots, i$ layers of molecules. If n_m is the maximum number of molecules that are adsorbed on the first layer, then

$$S_0 = \sigma_0 (n_m - n_1),$$
$$S_1 = \sigma_0 (n_1 - n_2), \qquad (1)$$
$$S_2 = \sigma_0 (n_2 - n_3), \text{ etc.},$$

where σ_0 is the area per molecule.

At equilibrium the rate of condensation must equal the rate of evaporation for each layer. Hence for the first layer,

$$a_1 p (n_m - n_1) = b_1 (n_1 - z n_2) e^{-E_1/RT}, \qquad (2)$$

where p is the pressure, E_1 the heat of adsorption in the first layer, and a, b, and z are constants. Analogous equations can be written for the higher layers.

The left-hand side of the equation shows that the rate of condensation is proportional to the pressure and the number of available sites. The right-hand side says that the rate of evaporation is proportional to the total number of molecules on that layer minus a certain effect produced by the molecules that are covered by two or more layers. The BET theory assumes $z = 1$, i.e., the molecules under a layer or two of molecules cannot evaporate through the superimposed layer. Hüttig, on the other hand (1948, p. 177), uses the other extreme that $z = 0$; hence he assumes that the molecules covered by second and higher layers still play their role as freely as if the higher layers were not present. Probably the true situation is somewhere between these extremes, but in the surface-area evaluation it is a second-order correction on the absolute value, as will be indicated below.

We will develop the BET isotherm following Brunauer (1943) and then compare the BET and Hüttig equations. The Hüttig equation may be derived in an exactly analogous manner from this point.

If we now multiply equation (2) by σ_0 and assume that z equals unity, we obtain:

$$a_1 p S_0 = b_1 S_1 e^{-E_1/RT}$$
$$\vdots \qquad \vdots$$
$$a_i p S_{i-1} = b_i S_i e^{-E_i/RT}. \qquad (3)$$

Since the total surface of the adsorbent is

$$A = \sum_{i=0}^{i=\infty} S_i \qquad (4)$$

and the total volume adsorbed is

$$v = v_0 \sum_{i=0}^{i=\infty} iS_i , \qquad (5)$$

where v_0 is the volume adsorbed per square centimeter by the surface when it is covered by a monolayer, it follows that

$$\frac{v}{A\,v_0} = \frac{v}{v_m} = \frac{\sum_{i=0}^{i=\infty} iS_i}{\sum_{i=0}^{i=\infty} S_i} , \qquad (6)$$

where v_m is the volume of gas adsorbed when a monolayer is complete.

From the basic assumption of the multimolecular theory,

$$E_2 = E_3 = E_4 = \ldots = E_i = E_L , \quad (7)$$

where E_L is the heat of liquefaction; further,

$$\frac{b_2}{a_2} = \frac{b_3}{a_3} = \ldots = \frac{b_i}{a_i} = g , \qquad (8)$$

where g is some constant.

In these steps we have assumed that the forces binding molecules in the second and higher layers are simply those of liquefaction. This is reasonable in view of the short range of Van der Waals forces.

It is now convenient to express S_1, S_2, etc., in terms of S_0.

$$S_1 = yS_0 , \quad \text{where} \quad y = \frac{a_1}{b_1}\, p\, e^{E_1/RT} , \quad (9)$$

$$S_2 = xS_1 , \quad \text{where} \quad x = \frac{p}{g}\, e^{E_L/RT} , \quad (10)$$

$$S_3 = xS_2 = x^2S_1 , \qquad (11)$$

$$S_i = xS_{i-1} = x^{i-1}S_1 = yx^{i-1}S_0 = Cx^iS_0 ; \qquad (12)$$

therefore,

$$C = \frac{y}{x} = \frac{a_1 g}{b_1}\, e^{(E_1 - E_L)/RT} . \qquad (13)$$

Substituting in equation (6), we obtain

$$\frac{v}{v_m} = \frac{CS_0 \sum_{i=1}^{\infty} ix^i}{S_0\left[1 + C\sum_{i=1}^{\infty} x^i\right]} . \qquad (14)$$

But

$$\sum_{i=1}^{\infty} x^i = \frac{x}{1-x} . \qquad (15)$$

Also

$$\sum_{i=1}^{\infty} ix^i = x\,\frac{d}{dx}\sum_{i=1}^{\infty} x^i = \frac{x}{(1-x)^2} . \qquad (16)$$

Now, substituting equations (15) and (16) in equation (14), we have

$$\frac{v}{v_m} = \frac{cx}{(1-x)(1-x+cx)} . \qquad (17)$$

At $p = p_0$ on a free surface all layers must be filled; therefore,

$$x = 1 \quad \text{or} \quad \frac{p_0}{g}\, e^{E_L/RT} . \qquad (18)$$

Therefore,

$$x = \frac{p}{p_0} . \qquad (19)$$

Substituting equation (19) in equation (17), we obtain the isotherm equation,

$$v = \frac{v_m c p}{(p_0 - p)\,[1 + (c-1)(p/p_0)]} , \qquad (20)$$

which may be transformed to

$$\frac{p}{v(p_0 - p)} = \frac{1}{v_m c} + \frac{c-1}{v_m c}\,\frac{p}{p_0} . \qquad (21)$$

If $p/v(p_0 - p)$ is plotted against p/p_0, a straight line should result with a slope equal to $(c-1)/cv_m$ and an intercept $1/v_m c$. It is therefore possible to determine both v_m and c. The former gives the surface area and the latter the heat of adsorption in the first layer, since E_L is known. To a good approximation on most surfaces, c is large compared to unity; hence to 2 per cent the slope may be taken as the reciprocal of v_m. This assumption may easily be checked by measuring the intercept for a few cases.

Reasoning from the Hüttig assumption, one obtains in place of equation (21) the following equation:

$$\frac{p}{v}\left(1 + \frac{p}{p_0}\right) = \frac{p_0}{c\,v_m} + \frac{p}{v_m} .$$

In this equation a graphical plot of $p/v\,(1 + p/p_0)$ against p or p/p_0 is a straight line with a slope equal to $1/v_m$. The comparison of these isotherms is shown in figure 2.

Ross (1948) shows some adsorption isotherms for ethane on NaCl at liquid-oxygen temperatures. Recomputed into the Hüttig equation, a straight line is obtained over a much larger range of p/p_0 than the simplified BET equation gives. Ross states that the longer linearity provides greater precision for the method. His data also indicate that, under the conditions he used, the surface-area values lie within 10 per cent of each other.

Because Halsey has already demonstrated some unreality in one of the assumptions underlying the multimolecular theory (whether BET or Hüttig), it is not profitable to discuss the implications of these differences at length. However, it should be mentioned that mere adherence to linearity does not necessarily prove the correctness of the equation. Further, it has been noted that the Hüttig equation is not linear for all types of adsorbents (G. Halsey, personal communication). Representative BET plots for the adsorption of argon on deep-sea sediment samples at liquid-nitrogen temperatures are shown in figure 2. The Hüttig plots for the same data are plotted for comparison. Note the tendency of the Hüttig plot to curve at low p/p_0 values. Thus Ross's claim to greater precision is questionable for these conditions. Further, the fact that the BET equation is linear to lower values of p/p_0 would favor the retention of BET surface-area values to the Hüttig values. Table 3 shows the comparison of the surface areas obtained for these three samples by the two methods. Generally, in this case the Hüttig values lie about 20 per cent higher. Until more conclusive theoretical work is done to establish the best equation for determining absolute surface-area values, it is felt best to use the BET for comparison. In any case, the absolute values are probably correct to 20 per cent.

TABLE 3

COMPARISON OF SURFACE AREAS CALCULATED BY THE BET AND HÜTTIG EQUATIONS

Core Sample	Depth (Cm.)	Surface Area (M^2/Gm)	
		BET	Hüttig
A153-141	1,025	5.39	6.31
A152-137	523	25.6	31.6
C8-7	145	12.5	16.4

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USE OF SOIL MECHANICS DATA IN CORRELATION AND INTERPRETATION OF LAKE AGASSIZ SEDIMENTS[1]

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ABSTRACT

The purpose of this paper is to illustrate the value of the use of soil mechanics in geology by the correlation and interpretation of sediments from glacial Lake Agassiz on the basis of soil mechanics test data. Quantitative data on the soil mechanics properties of liquid limit, plastic limit, natural water content, relative water content, and preconsolidation stress on samples from test borings at Grand Forks and Fargo, North Dakota, and Crookston, Minnesota, are used in establishing five stratigraphic units within the lacustrine sediments. Correlation of these units among the three localities is evaluated by means of statistical correlation coefficients. In addition, profiles of preconsolidation stress indicate a period of surface drying that denotes a short interval of drainage of Lake Agassiz not heretofore reported. Thus in this case the use of soil mechanics data yields information not readily obtainable by the usual geologic methods. It is believed that soil mechanics data could be used to considerable advantage in many other geological problems.

INTRODUCTION

For many years geologists and soil engineers have studied the same earth materials from two viewpoints, using techniques developed to yield data valuable in the sciences of geology and soil mechanics. As in all such instances of overlapping fields, the initial diversity of approaches is desirable, but a time is reached when workers in each field can profit by studying and applying the contributions of those in the other. This situation exists today in the fields of geology and soil mechanics; yet geologists in general have failed to use the mass of data made available through recent advances in soil mechanics, and, conversely, there is a failure on the part of many soil engineers to use geological data effectively. The purpose of the present paper is to point out the value of soil mechanics information in geology.

A few papers in which soil mechanics methods are discussed have appeared in the geologic literature (Terzaghi, 1940; Jones, 1944; Skempton, 1944; Fisk, 1947). Apparently, no papers using soil mechanics data in detailed stratigraphic correlation or in the determination of a hiatus by the effects of surface drying on preconsolidation stress have been published.

This paper describes results of a study of the sediments of glacial Lake Agassiz by means of quantitative soil mechanics data from tests on boring and test-pit samples from three localities: Grand Forks, North Dakota; Crookston, Minnesota; and Fargo, North Dakota (see fig. 1). Data used in this investigation are from tests made in connection with studies of foundation conditions at sites for power-plant buildings at these places. The test pits and borings from which all samples were obtained were made in 1946 for the Northern States Power Company and the Otter Tail Power Company. Laboratory tests on the samples were performed in the Soil Mechanics Laboratory of the Technological Institute, Northwestern University. The results were analyzed from the standpoint of foundation engineering by the second author, who submitted reports to the power companies in 1946 and 1947. The present paper consists of a reworking and integration of these

[1] Manuscript received November 15, 1950.

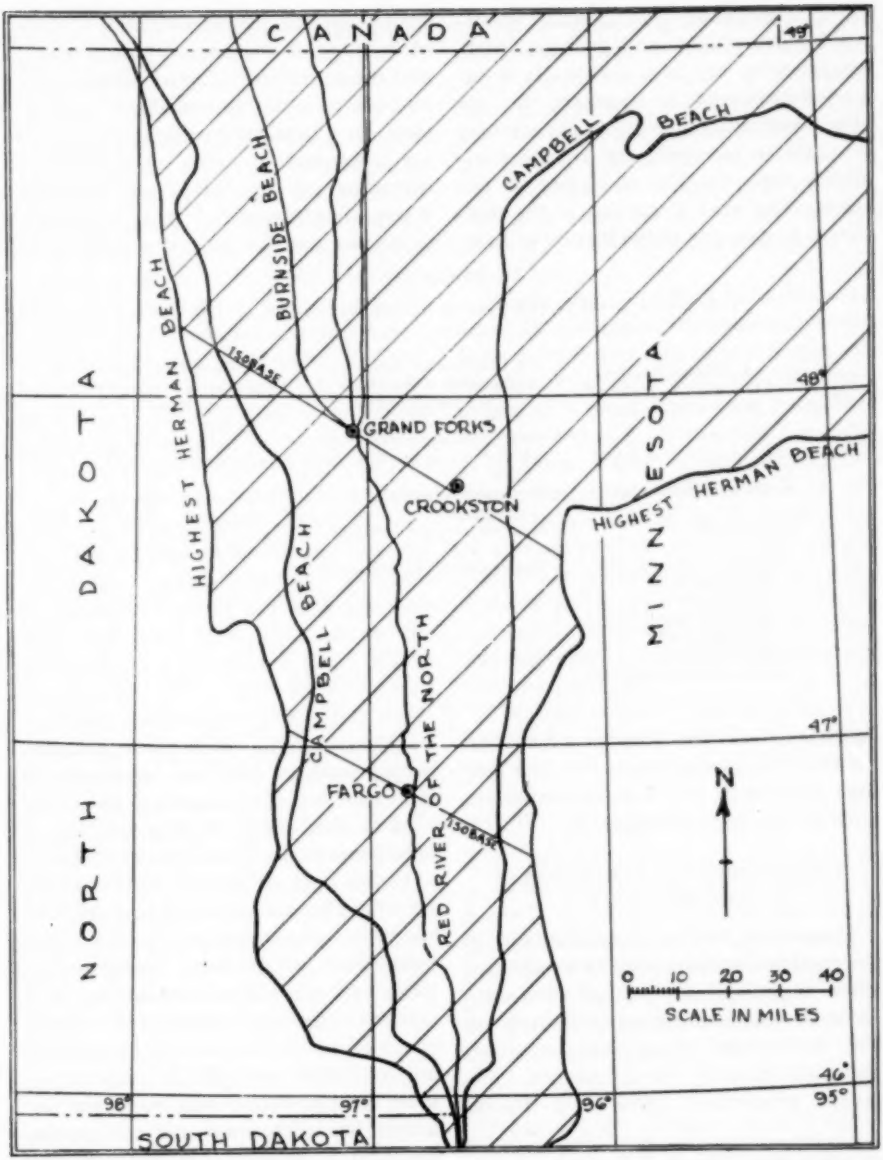


FIG. 1.—Index map

same engineering data, with stratigraphic correlation and geologic interpretation by the first author. It is intended primarily to illustrate the use of soil mechanics in geology. No attempt is made to integrate the results completely with the glacial history of the region. The brief discussion of the relation of test results to the history of Lake Agassiz is based on a rather cursory review of the geologic literature; no geologic field work was done in connection with the present investigation.

TABLE 1

TYPES OF QUANTITATIVE PHYSICAL PROPERTIES IN SOIL MECHANICS

A. Primary Properties (Dependent on Source and Depositional Conditions)	B. Intermediate Properties (Dependent on Both Basic Soil Material and Loading History)	C. Secondary Properties (Dependent on Postdepositional Stress History)
Specific gravity of grains	Natural water content: void ratio	Preconsolidation stress
Grain size distribution	Unconfined compressive strength	Relative water content (also called "water-plasticity ratio" or "liquidity index")
Grain shape	Shearing-strength relations	
Atterberg plasticity limits	Consolidation characteristics	
Liquid limit		
Plastic limit		
Plasticity index		
Shrinkage limit		
Permeability		
Compaction characteristics		

TYPES OF PHYSICAL PROPERTIES IN SOIL MECHANICS

Properties studied quantitatively in soil mechanics testing may be subdivided from a geological point of view into those that depend almost exclusively on the source and depositional environment of the soil[2] (herein termed "primary" properties), those that depend mainly on the postdepositional history of the deposit ("secondary" properties), and those that are affected considerably by both factors ("intermediate" properties). In general, the primary properties are measured in tests on completely disturbed or remolded soil samples, whereas intermediate and secondary properties are determined reliably only from tests on the best possible undisturbed samples that are essentially in the condition of the natural soil. This kind of distribution of properties tested in soil mechanics is indicated in table 1.

In this analysis of Lake Agassiz clays, only data for the following properties of the three groups have been used: (1) liquid limit, plastic limit, and plasticity index; (2) natural water content; and (3) preconsolidation stress and relative water content. Data for other properties are available and will be included in more comprehensive studies. In the following brief descriptions of the properties used, no attempt is made to describe the tests in detail. For such descriptions the reader is referred to the works of Casagrande and Fadum (1940), Skempton (1944), and Taylor (1948). The dis-

[2] In this paper the word "soil" is used in its soil mechanics connotation of all rock material at or near the surface that is nonlithified—i.e., not "bedrock"—rather than in the more restricted geologic usage with reference to a soil profile, etc., and excluding unaltered "rock," regardless of its state of lithification.

cussion of soil properties follows the grouping in table 1.

PRIMARY PROPERTIES

Liquid limit.—Liquid limit is the water content, expressed in percentage of dry weight, at which a remolded soil is just capable of resisting a measurable static shearing stress. In other words, it is a measure of the boundary between the plastic solid state and the viscous liquid state of a material. Determination of liquid limit was standardized by Casagrande (1932), who developed a device with which a careful worker can obtain reproducible results. In his method a remolded cake of the material is placed in a metal dish, and a standard tool is used to cut a tapered groove (8 mm. deep and 2 mm. wide at the bottom) across the center of the cake. The dish is mounted above an eccentric cam such that each turn of a crank causes the dish to drop a distance of 1 cm. onto a hard rubber block. The water content at which 25 blows, resulting from turning the crank two times per second, are sufficient to close the bottom of the groove for a distance of 1 cm. is defined as the liquid limit. This procedure is described in detail by Casagrande and Fadum (1944, pp. 19–20).

Although the factors controlling liquid limit are inadequately known, it apparently depends primarily upon the mineralogic composition of the material, plus such attributes of the individual particles as grain size and sorting. In particular, it seems to be closely related to the types and percentages of clay minerals, to the percentage of organic matter, and to the percentage of silt and coarser material present in the soil. Liquid limit is not influenced by the aggregate or mass properties of the sediment, such as stratification, porosity, or other grain-to-grain relationships, because the material is remolded before being tested. In the case of the Lake Agassiz clays now under consideration and in other sedimentary clays, the liquid limit reflects the composition of the material as it was deposited, plus the results of any changes in mineralogy which may have occurred subsequent to deposition.

Plastic limit.—"The plastic limit of a soil is defined as the water content (expressed in per cent of dry weight) at which a soil will crumble when rolled into a thread 3 mm. (1/8″) in diameter" (Casagrande and Fadum, 1940, p. 19). It is thus a measure of the boundary between the plastic solid state and the friable (or semielastic, brittle) solid state of a material. Like liquid limit, plastic limit is a mass property of the remolded material and depends upon the mineralogy and grain size rather than upon the aggregate properties of the undisturbed sample.

Plasticity index.—Plasticity index (I_p) is the difference between the liquid-limit (w_l) and the plastic-limit (w_p) water contents—i.e.,

$$I_p = w_l - w_p .$$

It is, therefore, a measure of the range in water content over which a material exists in a plastic solid state. The liquid limit and the plasticity index together provide a fairly complete measure of plasticity characteristics. As such, they are used effectively for classification of clay soils (Casagrande, 1947).

INTERMEDIATE PROPERTIES

Natural water content.—Natural water content, expressed in percentage of dry weight, is the ratio of the loss in weight sustained by a sample in drying at 105° C. to the weight of the dried sample. In many sedimentary clays the natural

water content immediately after deposition is equal to or slightly greater than the liquid limit of the material. With subsequent compaction, water is squeezed out, and the natural water content decreases. Unlike the Atterberg limits, natural water content is a mass property of the clay, depending upon such grain-to-grain relationships as size and configuration of voids, resulting from both depositional conditions and loading history, as well as upon mineralogic composition and other properties of individual grains. Although its magnitude may be different in two areas having a similar lithologic succession, its relative variation within each area is generally correlative with the lithologic changes. Theoretically, it should decrease downward, owing to increasing compaction downward; but in most cases, at least at shallow depths, this trend is subordinate to variations associated with variations in lithology. Changes in natural water content with depth are relatively independent of the position of the water table, because water is held in the voids of the clay by capillary forces and by molecular forces associated with the structure of the various clay minerals, and hence does not change markedly at the water table. It should be pointed out, however, that the precise relationships between natural water content and the more fundamental properties of clays are inadequately known.

SECONDARY PROPERTIES

Preconsolidation stress.—The consolidation test is a measure of the relation between stress and volume change in a confined specimen of soil. It is one of the basic physical tests of soil mechanics, the results being used in predicting the magnitude and rate of settlement of buildings founded on clays. Consolidation test data are compiled in compression diagrams, in which stress (logarithmic scale) is plotted against volume (arithmetic scale), usually expressed as void ratio—i.e., ratio of volume of voids to volume of solids. The initial portion of such compression diagrams (see fig. 2) shows a small decrease in void ratio (hence in total volume) per unit increase in compressive stress. With continued increase in stress, the rate of decrease in volume is increasingly larger, until it becomes constant (in terms of stress plotted to a logarithmic scale) at a relatively high volume change per unit increase in stress, producing the characteristic straight, or slightly concave upward, portion of the curve. The similarity in shape between the initial laboratory curve (segment $\overline{AB}$, fig. 2) and the curve ($\overline{EF}$) obtained by reloading following the release of load in the middle of the test ($\overline{CD}$) has led to the concept that the shape of the initial laboratory curve is the result of previous loading in the history of the soil—i.e., that the initial laboratory curve is really a reloading curve following rebound from initial natural loading. On this basis the "preconsolidation stress," or maximum stress that has affected the soil prior to laboratory sample study and prior to field consolidation, resulting from building loads, etc., can be estimated, as discussed more fully below.

Undisturbed clay samples for consolidation tests are removed from their natural site by hand tools in test pits or by large-diameter (5-inch) sampling devices in bore holes. Below the zone of weathering they are almost invariably saturated. The methods of sampling and the very large possible capillary forces in the saturated clay when exposed to air practically insure a constant volume

of the sample if drying is prevented. Hence the laboratory specimen initially has essentially the same volume that it had in nature. However, the clay has undergone some disturbance during the sampling operation that weakens its structure and lowers its equilibrium curve of stress versus volume change, similar to the displacement of the laboratory recompression curve $\overline{FG}$ below the initial laboratory curve $\overline{BC}$ in figure 2. Knowledge of the displacement of the compression curve due to sample disturbance between tests and of the shape of the laboratory rebound curve would permit determination of the maximum stress used in the first test (point C) from data obtained in the second test. Similarly, knowledge of these two factors would permit determination of the natural preconsolidation stress from the initial laboratory test data.

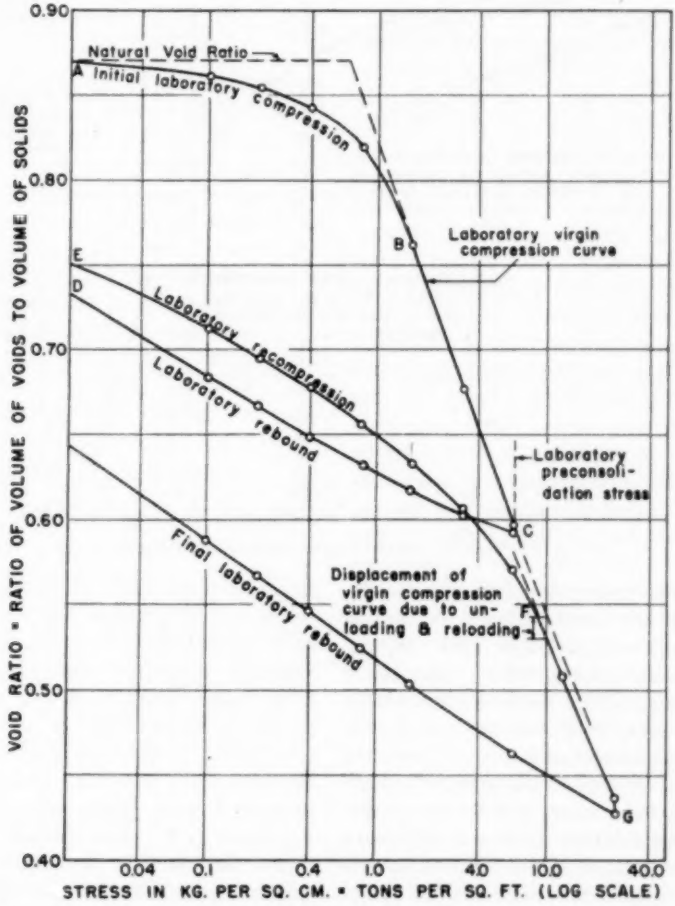


FIG. 2.—Typical compression diagram (actual test on clay sample from Chicago subway)

The range of possible values of preconsolidation stress determined in a laboratory consolidation test is shown in figure 3. The preconsolidation stress cannot be less than the value on the virgin compression curve at the natural void ratio (*H*), and it cannot be greater than a value corresponding to a full rebound prior to initial laboratory loading (*J*). The most probable value of preconsolidation stress within this range is determined by a method proposed by Casagrande (1936); the method is indicated graphically in figure 3, and the reader is referred to Casagrande's original work for a more detailed treatment.

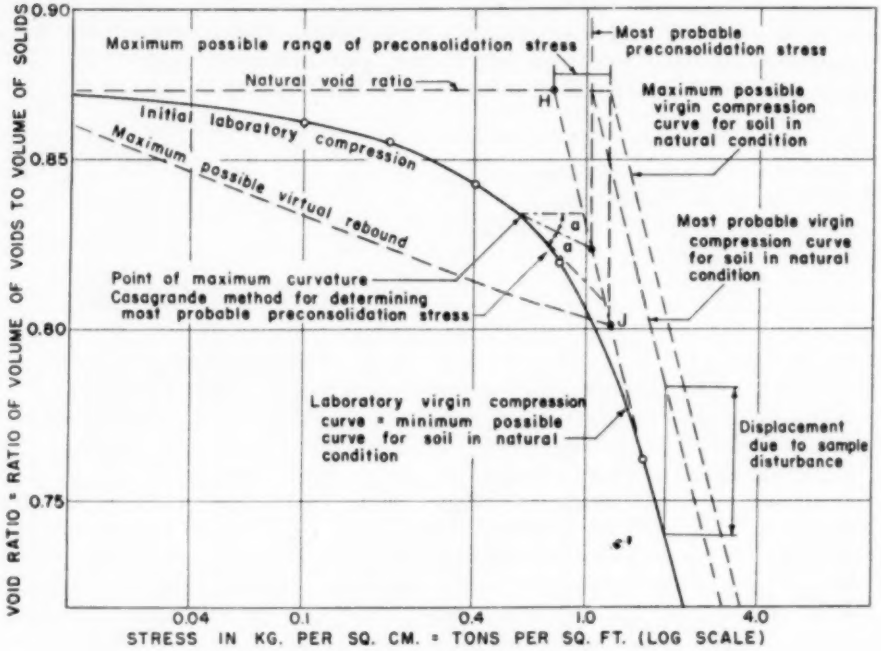

VOID RATIO = RATIO OF VOLUME OF VOIDS TO VOLUME OF SOLIDS
0.90
0.85
0.80
0.75
Maximum possible range of preconsolidation stress
Most probable preconsolidation stress
Natural void ratio
H
Initial laboratory compression
Maximum possible virtual rebound
Maximum possible virgin compression curve for soil in natural condition
Most probable virgin compression curve for soil in natural condition
a
a
Point of maximum curvature
Casagrande method for determining most probable preconsolidation stress
J
Laboratory virgin compression curve = minimum possible curve for soil in natural condition
Displacement due to sample disturbance
σ'
0.04
0.1
0.4
1.0
4.0
STRESS IN KG. PER SQ. CM. = TONS PER SQ. FT. (LOG SCALE)

FIG. 3.—Relation of preconsolidation stress to laboratory test curve

Preconsolidation stress is a significant property because it can be used in predicting, with a fair degree of accuracy, the maximum stress that has acted on the clay for any appreciable period of time. The stress may have been caused by an overburden of soil, glacial ice, buildings, etc., or by capillary forces resulting from desiccation at a ground surface. The indications of a drying surface are particularly consistent in a series of tests made at various depths below the desiccation surface. In this case preconsolidation stress almost invariably decreases consistently from large values near the drying surface to values corresponding to the existing overburden stresses at greater depths. Because of the reliability with which the preconsolidation stress reflects the loading history of a soil deposit, it is believed that it may become a valuable quantitative tool for geologists, particularly in the location of unconformi-

ties and in the calculation of ice thickness and amount of erosion.

Relative water content.—Relative water content (w_r), also called "water-plasticity ratio" or "liquidity index," is defined as the ratio of the difference between natural water content (w_n) and plastic limit to the difference between liquid limit and plastic limit (hence, to plasticity index):

$$w_r \text{ (in per cent)} = \frac{w_n - w_p}{w_l - w_p} \times 100 = \frac{w_n - w_p}{I_p} \times 100.$$

Thus relative water content is a measure of the relation of natural water content to the liquid and plastic limits of the soil. For this reason it reflects strongly the loading history of a soil. For example, two immediately adjacent samples of soil may have natural water contents of 25 and 45 per cent, the difference being due to differences in lithology, but the relative water contents might both be 50 per cent. The difference between the two soils is shown by the liquid and plastic limits, and the similarity in loading history is shown by the relative water contents. Without plasticity limits or fairly definite knowledge of the lithology of a soil deposit, knowledge of variations in natural water content has limited quantitative significance. However, because lithological changes often have effects on the natural water content that are large compared with other effects, natural water content variations are frequently used for preliminary differentiation between soil strata.

THE LAKE AGASSIZ DATA

Data used in the present discussion of the Lake Agassiz sediments comprise (1) liquid limit, (2) plastic limit, (3) natural water content, (4) plasticity index, (5) relative water content, and (6) preconsolidation stress, all of which are described in the preceding section, plus (7) lithology and (8) structure. In this case, "lithology" consists of qualitative observations of the megascopic characteristics of the clays, including the nature of the bedding, made in the field by the drillers and refined in the laboratory by the engineers testing the individual samples.

This paper is based on information obtained from three sample points, located as follows: (1) Grand Forks, North Dakota, Northern States Power Company plant on the west bank of the Red River of the North, between Kittson and Demers avenues, Grand Forks; (2) Crookston, Minnesota, Otter Tail Power company plant on the west bank of Red Lake River at the northwest edge of the town of Crookston; and (3) Fargo, North Dakota, Northern States Power Company proposed plant site on the west bank of the Red River of the North, at Fifth Avenue North and Elm Street, Fargo. At Grand Forks, 117 undisturbed samples were taken from one test pit and seven bore holes; at Crookston 14 undisturbed samples and 99 disturbed samples (sealed for natural water content tests) were taken from one test pit and six bore holes; and at Fargo 32 undisturbed samples and 86 disturbed samples were taken from five bore holes.

The study of these samples yielded the eight types of information listed above, plus certain compressive strength data not used in the present discussion. Unfortunately, however, all the tests were not made on each of the samples, and for this reason the completeness of the information varies among the three localities, the information from Grand Forks being fairly complete, from Fargo

TABLE 2

SUMMARY OF AVAILABLE TYPES OF INFORMATION* ON LAKE AGASSIZ CLAYS

	Grand Forks	Crookston	Fargo
Measured data:			
1. Liquid limit	××	x	××
2. Plastic limit	××	x	××
3. Natural water content	××	××	××
4. Preconsolidation stress	××	××	—
Derived parameters:			
1. Plasticity index	××	x	××
2. Relative water content	××	x	××
Qualitative observations:			
1. Lithology	××	××	××
2. Structure	x	—	—

* ××=Fairly complete information; x=incomplete information; —=no information.

almost as complete, and from Crookston quite incomplete. A summary of the types of information available at the three localities is given in table 2.

The actual test data for each of the localities, plotted against depth, are summarized graphically in figure 4, which includes all information relevant to the present discussion except lithology (table 3) and structure.

STRATIGRAPHY OF THE LAKE AGASSIZ CLAYS

THE FIVE STRATIGRAPHIC UNITS

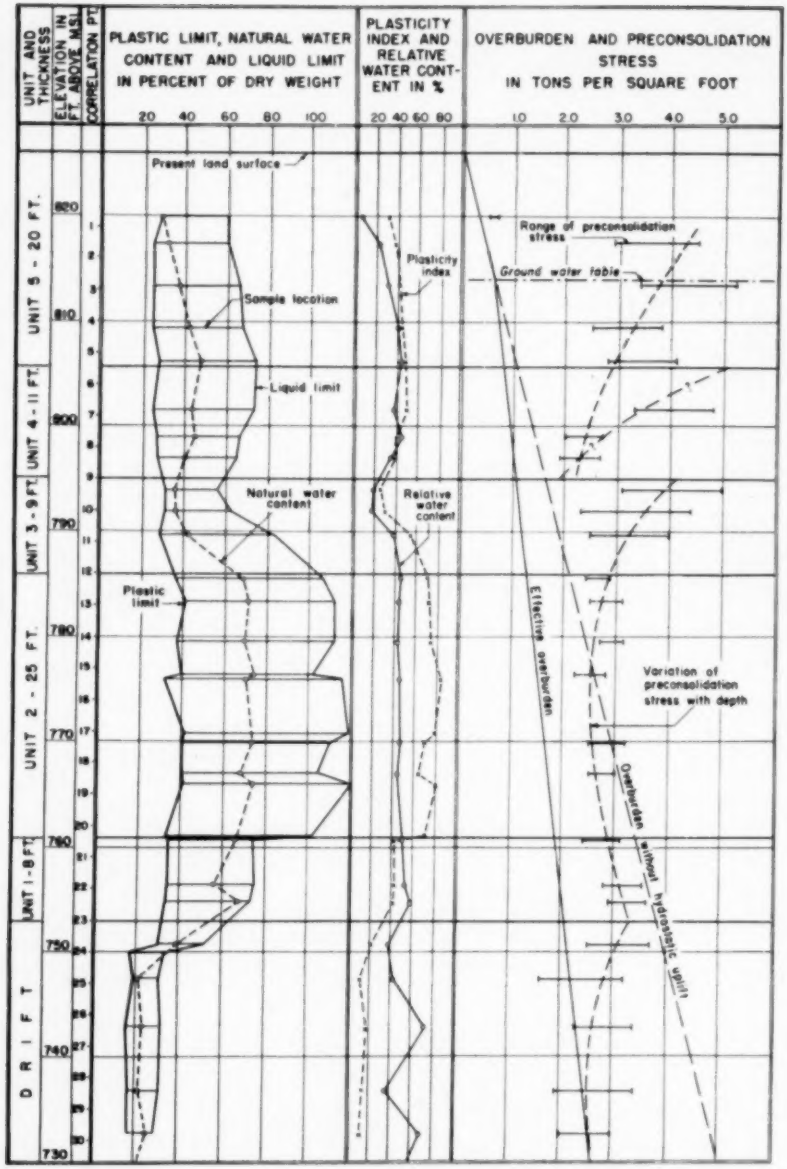
A glance at the graphical presentation of data from Grand Forks and Fargo

TABLE 3

GENERALIZED LITHOLOGIC DESCRIPTIONS

Unit	Grand Forks, N.D.	Crookston, Minn.	Fargo, N.D.	Fargo, N.D., by Laird
5	Brown clay and silt, stratified 20 ft.	Brown clay and silt, stratified 18 ft.	Yellow silt and clay, nonstratified 15 ft.	Buff silt and clay 19 ft.
	unconformity			unconformity
4	Dark-blue clay, mostly nonstratified 11 ft.	Blue clay, nonstratified 24 ft.	Dark brown-green clay, with interbedded sand and silt, fossil shell fragments, and organic matter in lower half 12 ft.	
	old drying surface	old drying surface	old drying surface	
3	Dark-blue clay, mostly nonstratified, but with some interbedded silt at top 9 ft.	Blue clay, nonstratified, with isolated pebbles 22 ft.	Yellow silt, friable 3 ft.	Gray or blue clay 72–83 ft.
2	Dark-blue or black clay, nonstratified, many slickenside surfaces, high liquid limit and high natural water content 25 ft.	Blue clay, nonstratified, with isolated pebbles, high liquid limit, and high natural water content 18 ft.	Blue-black clay, with calcareous concretions, nonstratified, high liquid limit and natural water content 20 ft.	
1	Dark-blue clay with calcareous concretions, nonstratified 8 ft.	Blue clay, nonstratified, with isolated pebbles 5 ft.	Dark brown-green clay 7 ft. Blue-black clay, with calcareous concretions 28 ft.	
	unconformity Drift 73+ feet	unconformity Drift 6+ feet	unconformity Drift 3+ feet	unconformity Drift 170 feet

GRAND FORKS



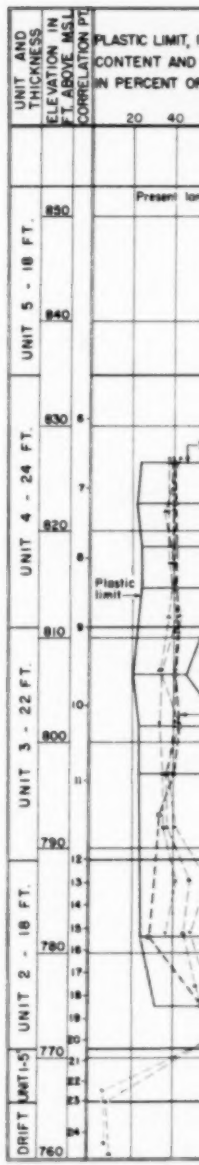
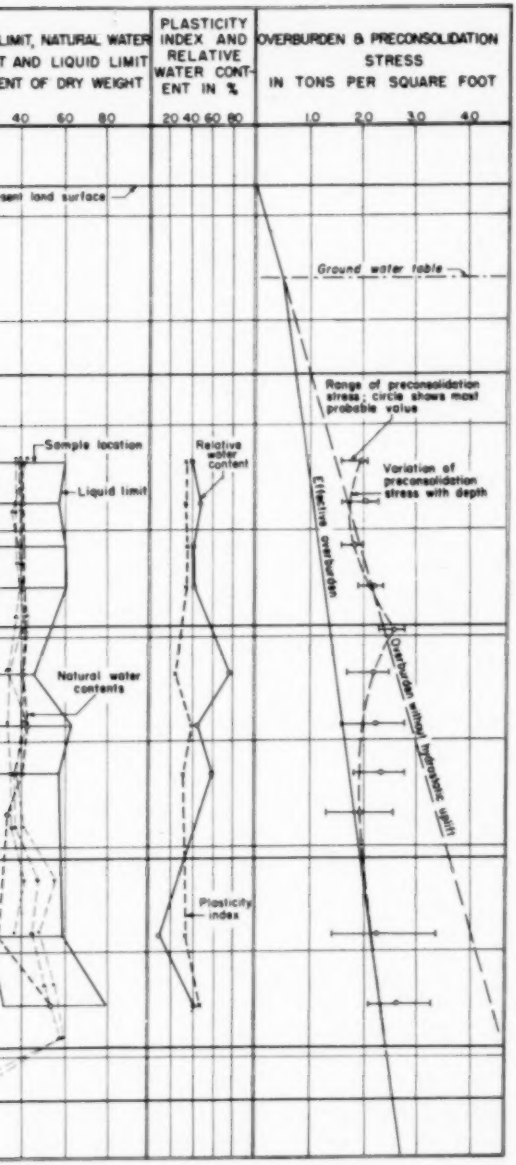
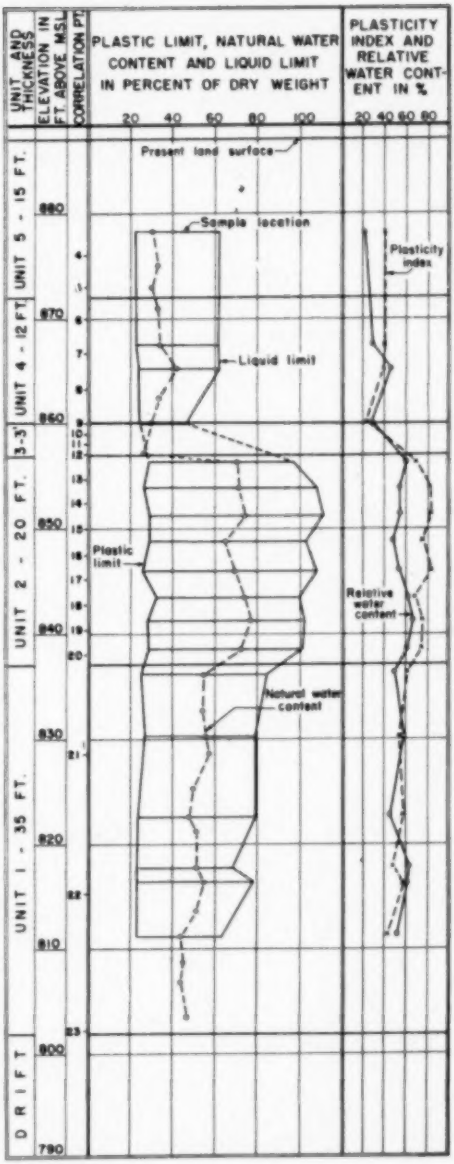


FIG. 4.—Grand Forks, Crookston, and Fargo soil

CROOKSTON



FARGO



go soil mechanics data plotted against depth

(fig. 4) gives one the impression that there are stratigraphic units common to both localities. A study of the data confirms the impression and indicates, further, that the same units are present at Crookston, though not quite so distinctly. Detailed study of these data has resulted in subdivision of the lacustrine clays overlying the till into five stratigraphic units as shown in table 4. It should be noted that these divisions are rock units, not time units, though they are no doubt essentially contemporaneous throughout the area. Noteworthy also is the fact that units 1, 2, 3, and 4 appear almost homogeneous under the usual qualitative inspection and have quite naturally been grouped together as one lithologic unit in previous work. Subdivision into these four units is made possible mainly by the break in the preconsolidation stress curves between units 3 and 4, indicating the presence of an old drying surface, and by the characteristic high liquid limit and natural water content of unit 2. In the light of these data, however, a re-examination of the sediments shows megascopic lithologic differences between the units, such as the black color of unit 2 and the shells immediately above the old drying surface.

TABLE 4

STRATIGRAPHIC UNITS OF THE LACUSTRINE CLAYS

Unit	Description	Thickness (Feet)
	(Surface)	
5	Brown silt and clay, stratified	12–20
	(Local unconformity)	
4	Blue, brown, or dark-green clay, mostly nonstratified, with some organic matter and shell fragments	11–24
	(Old drying surface)	
3	Blue clay, nonstratified	3–22
2	Dark-blue or black clay, nonstratified, with very high liquid limit and natural water content	18–25
1	Blue clay, nonstratified, with silt lenses, calcareous concretions, and isolated pebbles	5–35
	(Unconformity)	
	Glacial drift (Wisconsin)	

Grand Forks, where the data are most complete and where differences are most apparent, is the type locality for the five stratigraphic units. The wide range of evidence used in defining the units is indicated in table 5, which comprises criteria used in locating the upper contact of each unit at Grand Forks, Crookston, and Fargo. The criteria given for a contact are listed in approximate order of importance—for example, the most striking change from unit 3 to unit 2 at Grand Forks is the sudden increase in liquid limit, and of successively less obvious nature are changes in natural water content and lithology. As shown in the table, the five contacts at Grand Forks are all based on three or more lines of evidence and the occurrence of such changes at the several definite horizons gives strong support to the validity of the fivefold division.

The subdivisions of lithology and thickness of the units at each of the three localities are made according to the criteria listed in table 5 and shown graphically in figure 4.

THE OLD DRYING SURFACE

Probably the most significant feature in the test data is the break in the preconsolidation stress curve above unit 3 at Grand Forks and Crookston (fig. 4), for this feature indicates an old drying surface within a series of clays thought to have been formed during a single depositional stage of Lake Agassiz (see p. 177). As previously stated, the effect of drying at a land surface is compaction

of the surface clays, and this compaction is reflected in the preconsolidation stresses. Characteristically, the preconsolidation stresses decrease downward from a value of 4 tons per square foot or greater at the drying surface to a value at a depth of 10 to 30 feet beneath the surface equal to the overburden pressure; below this point they increase in proportion to the increase in overburden. The subsurface highs of preconsolidation stress at Grand Forks and Crookston are of this characteristic configuration and undoubtedly indicate a period of surface drying following the deposition of unit 3.

Associated with the change of preconsolidation stress are decreases in liquid limit at Grand Forks and Crookston and in relative water content at Grand Forks. Similar decreases in liquid limit and relative water content occur at Fargo, where data on preconsolidation stress are lacking, and provide means of determining the position of the drying surface at this locality. Such changes in liquid limit and relative water content are expectable at a drying surface and give support to the evidence from the preconsolidation stress data. The presence of shell fragments in the material immediately overlying this horizon at Fargo is also in accordance with this interpretation.

There is a suggestion of a break in the preconsolidation stress curve at the

TABLE 5

CRITERIA USED IN DETERMINING TOPS OF STRATIGRAPHIC UNITS, LISTED IN ORDER OF IMPORTANCE

Unit	Grand Forks, N.D.	Crookston, Minn.	Fargo, N.D.
5	(Ground surface)	(Ground surface)	(Ground surface)
4	1. Lithology 2. Structure 3. Preconsolidation stress (?)	1. Lithology	1. Lithology
3	1. Preconsolidation stress 2. Liquid limit 3. Relative water content 4. Plasticity index 5. Natural water content	1. Preconsolidation stress 2. Liquid limit	1. Liquid limit 2. Relative water content 3. Plasticity index 4. Lithology
2	1. Liquid limit 2. Natural water content 3. Lithology	1. Natural water content 2. Liquid limit (?)	1. Liquid limit 2. Lithology 3. Natural water content 4. Relative water content 5. Plasticity index
1	1. Liquid limit 2. Plasticity index 3. Lithology (?)	1. Natural water content	1. Liquid limit 2. Natural water content 3. Lithology 4. Plasticity index 5. Relative water content
Drift	1. Lithology 2. Liquid limit 3. Plastic limit 4. Preconsolidation stress 5. Plasticity index	1. Lithology	1. Lithology

known unconformity above unit 4 at Grand Forks, as shown by the alternate curve in figure 4, but the data points in the upper part of unit 4 are too few to permit interpretation with the assurance warranted for the break at the top of unit 3. Although the data at Crookston do not extend to the top of unit 4, there is a tendency for the preconsolidation stress curve at the highest data point to increase upward, in the manner expectable below an unconformity. A definite high point in the preconsolidation stress curve also occurs at the unconformity above the till underlying the lacustrine clays at Grand Forks.

CORRELATION BETWEEN SAMPLE LOCALITIES

The persistence of the test characteristics of the units from one locality to the next is remarkable, but correlation of units 1 to 4 on the basis of lithology alone would have been practically impossible. A study of figure 4 shows that the relative changes between units are the same at each of the localities, even though the magnitudes of the different test results are not identical at the three localities. For example, unit 2, everywhere of relatively high liquid limit, is everywhere overlain by a layer of moderate liquid limit (unit 3), the upper portion of which has been subjected to the compactive effects of surface drying. It is this succession of characteristics that gives strength to the correlation—strength that would be lacking if correlation were based upon only one distinctive bed.

As in most stratigraphic correlations, the thickness varies between localities, although the succession of beds is the same at each of the localities. Changes in thickness between Grand Forks and Fargo are minor, and correlation between these two localities is better than that between either of them and Crookston. The relative weakness of the Crookston correlation, in addition to being partially due to incomplete data, probably results from the fact that Crookston is closer to the edge of Lake Agassiz than either Grand Forks or Fargo, which are both near the center. In particular, unit 2 at Crookston is thinner and farther below the old drying surface (owing to a considerably thicker unit 3) than at the other two localities. Although the same relative changes occur above and below this unit as at the other localities, there is a possibility that the unit is not continuous with the Grand Forks–Fargo unit 2 and really represents a lens of similar material at approximately the same horizon. In view of the uniformity of deposition expectable in such a large lake, however, the layer of high liquid limit at Crookston is correlated with the Grand Forks unit 2.

Variations within each locality are similar to those between the localities; each of the units can be recognized in all borings on which sufficient data are available, but the thicknesses of the units at each locality vary among the holes drilled within the few hundred square feet included in that locality. The data shown in figure 4 for each locality are from the particular boring or test pit for which the greatest amount of sample information is available. In addition, natural water content values from four borings at Crookston are superposed on the main Crookston data. In the absence of information on other properties, these data help fill the gap in liquid-limit sample points at Crookston. The consistent change in natural water content between elevations 787 and 792 in these holes is used as the basis for locating the top of unit 2, resulting in a

thicker unit 2 than could be interpreted from the liquid-limit information.

QUANTITATIVE EVALUATION OF THE STRATIGRAPHIC CORRELATION

Method.—In order to check the correlation as a whole and to evaluate the effectiveness of each of the quantitative test properties in correlation, scatter diagrams were plotted and statistical correlation coefficients computed for the stratigraphic variation of each of the properties between Grand Forks and Fargo and of one of the properties between Grand Forks and Crookston. Arbitrary correlation points, spaced at 3-foot intervals (the average distance between sample points) above and below the old drying surface, were established on the Grand Forks section and numbered consecutively from top to bottom (see fig. 4). Points located at the corresponding positions within the units at Fargo and Crookston were determined and numbered accordingly. For example, correlation point 11 at Grand Forks is within unit 3 and is one-third of the way up from the base of that unit; hence both point 11 at Fargo and point 11 at Crookston are located one-third of the way up from the base of unit 3. By this method a complete set of correlation points, located in an unbiased manner but dependent upon the previously established correlation of stratigraphic units, is available to serve as a basis for testing the reliability of the correlation.

At each correlation point, pairs of values of the test properties for which information is complete were read and plotted on the scatter diagrams shown in figure 5. Thus the value of liquid limit at correlation point 11 at Grand Forks is 88 per cent and the corresponding value at Fargo is 74 per cent; these values were used as co-ordinates in plotting one of the points on the Grand Forks–Fargo liquid limit scatter diagram. If the relative changes, on a thickness basis, between and within all the units at Grand Forks and Fargo were identical for a given property (regardless of the absolute magnitude of that property), the points would all fall on a straight line. The degree that the plotted points approach such a straight-line relationship is a measure of (1) the uniformity of the rock units, in terms of the property in question, and (2) the accuracy of the correlation. The line drawn through each set of points in figure 5 is the best fit, in the sense of least squares, and thus represents the best estimate of the relationship between values at the two localities.

A quantitative measure of the approach of such data to a linear relationship is provided by the correlation coefficient (r) of statistics. This parameter is computed with little difficulty by the so-called "product-moment" method; its computation and significance are discussed in nearly all standard books on statistics (see Mills, 1938, pp. 349–359). The correlation coefficient varies from 0 to 1 as the two arguments vary from absolute independence to perfect linear correlation.

Results.—Results of the evaluation of the Grand Forks–Fargo correlation by the above methods are shown graphically in the scatter diagrams of figure 5 and numerically in the list of correlation coefficients given in table 6. The exceptionally high values of the correlation coefficients for natural water content, liquid limit, plasticity index, and plastic limit give unquestionable support to the correlation as a whole between the two localities—that is, to correlation of the entire succession of units, not just of the units individually.

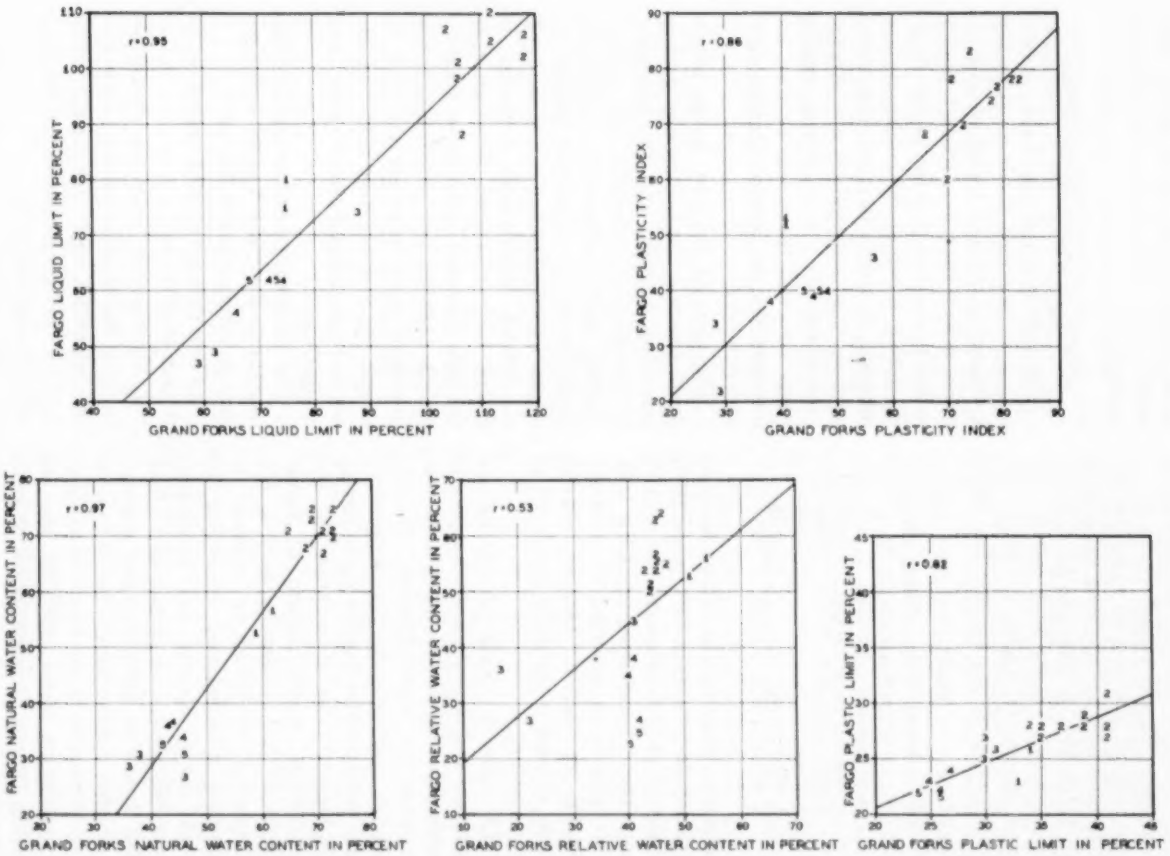

r=0.95
FARGO LIQUID LIMIT IN PERCENT
GRAND FORKS LIQUID LIMIT IN PERCENT
r=0.86
FARGO PLASTICITY INDEX
GRAND FORKS PLASTICITY INDEX
r=0.97
FARGO NATURAL WATER CONTENT IN PERCENT
GRAND FORKS NATURAL WATER CONTENT IN PERCENT
r=0.53
FARGO RELATIVE WATER CONTENT IN PERCENT
GRAND FORKS RELATIVE WATER CONTENT IN PERCENT
r=0.82
FARGO PLASTIC LIMIT IN PERCENT
GRAND FORKS PLASTIC LIMIT IN PERCENT

FIG. 5.—Grand Forks–Fargo scatter diagrams. Correlation coefficient = r. Numbers refer to stratigraphic units

The relative values of the correlation coefficients for the Grand Forks–Fargo data indicate that natural water content and liquid limit are the most valuable of the properties in stratigraphic correlation and that plastic limit and the derived parameter, plasticity index, are next in value. It is gratifying that natural water content ranks so high in correlation value, because this property is relatively easy to measure. The relative water content correlates only fairly well. This should be expected in a stratigraphic correlation, because relative water content is primarily a measure of stress history.

TABLE 6

GRAND FORKS—FARGO CORRELATION COEFFICIENTS

Method	r
1. Natural water content	0.97
2. Liquid limit	0.95
3. Plasticity index	0.86
4. Plastic limit	0.82
5. Relative water content	0.53

The data at Crookston are too incomplete and the correlation too poor to merit reproduction of the scatter diagrams in this paper. However, the correlation coefficient for the supplementary natural water content curve having the highest value at elevation 787 and the natural water content at Grand Forks is 0.53, indicating only a fair correlation. It would be incorrect to compute a correlation coefficient for liquid limit, because the liquid-limit data indicate a much thinner unit 2 than that based on natural water content. The most definite feature in the Crookston correlation is the break in the preconsolidation stress curve at the old drying surface. It is impossible to evaluate such a correlation by computation of a correlation coefficient for the depth variation in preconsolidation stress, because, except at the drying surface, this factor varies with depth, not with the stratigraphy.

SUMMARY OF METHOD OF CORRELATION

The correlation of the Lake Agassiz sediments described above is essentially a subsurface method based on quantitative sample analysis. Although similar methods could be applied to the study of certain types of data from surface sampling, the particular properties measured in this investigation are affected considerably by surface drying, weathering, etc., and thus could be used for interpretation of surface samples only with difficulty.

The plots of the test values versus depth resemble the electric logs used in petroleum geology, and the principles of stratigraphic correlation are similar. Indeed, such correlation coefficients could be used in petroleum geology to evaluate correlation and to test the merit of various geophysical methods. It should be remembered, however, that the data discussed in this paper are from sample analyses, not geophysical measurements; they represent the type of sample log that could be made, for example, by plotting mean grain size against depth.

The complete procedure used in correlation of the Lake Agassiz sediments may be summarized as follows:

1. Test data are plotted against depth at each locality.

2. Stratigraphic units are chosen by study of the depth variation of the several properties, such that greater variations of the properties occur between units than within units.

3. The criteria used in determining the upper contact of each of the units at each locality are listed, as a qualita-

tive indication of the integrity of the units.

4. Arbitrary correlation points at corresponding stratigraphic horizons are established at the three localities.

5. Scatter diagrams are plotted to show graphically the degree of correlation of the various test data between localities.

6. Correlation coefficients are computed to give a quantitative evaluation of (*a*) the correlation as a whole and (*b*) the relative merit of the various properties in correlation.

MINERALOGY AND GRAIN SIZE OF LAKE AGASSIZ CLAYS

As a matter of interest, the incomplete data on the mineralogy and grain size of the Lake Agassiz clays are discussed briefly in the following paragraphs.

Soil mechanics classification diagrams of liquid limit versus plasticity index for each of the three localities, using the data of figure 4, are given in figure 6.[3] The two dashed lines on each of the diagrams delimit the area that, according to past experience, includes most of the inorganic clays (Casagrande, 1947). Nearly all the Lake Agassiz points fall within this area, although the sediment may include some organic matter. The points representing unit 2 are grouped in the upper right portion of the Grand

[3] The plot of liquid limit versus plasticity index, instead of the plot of liquid limit versus plastic limit, is used here in conformity with soil mechanics practice.

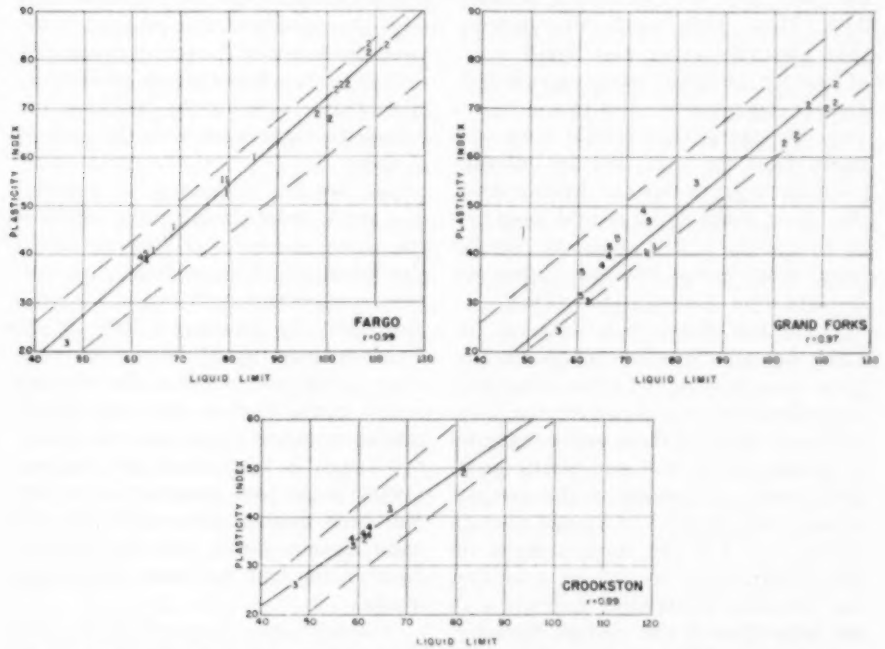


FIG. 6.—Plasticity classification diagrams for Grand Forks, Crookston, and Fargo clay samples. Dashed lines delimit area of most inorganic clays; solid line is best fit in sense of least squares; numbers refer to stratigraphic units; r = correlation coefficient.

Forks and Fargo diagrams, in the high liquid-limit range characteristic of montmorillonite clays, strongly suggesting that this unit contains considerable montmorillonite. Unit 2 at Crookston and the other units at all three localities may also contain montmorillonite, their lower liquid limits and plastic limits being due to differences in grain size distribution or to greater amounts of admixed quartz particles. No other mineralogic data are available to check these inferences.

The solid line in each of the diagrams of figure 5 represents the best fit in the sense of least squares, thus indicating the most probable variation of plasticity index with liquid limit. The correlation coefficient for the Grand Forks data is 0.97, for Fargo 0.99, and for Crookston 0.99. These high coefficients indicate that plasticity index and liquid limit, at least for the Lake Agassiz clays tested, are not independent. And this is equivalent to saying that plastic limit and liquid limit for the clays are interdependent properties or are dependent on the same more fundamental property or properties of the material. Similar linear relationships have been observed in clays from many other localities.

Grain size distribution data on the Lake Agassiz clays are far too incomplete to warrant reproduction here. Only seven hydrometer analyses are available, and only three of these are on samples of known liquid limit and plastic limit. The median diameters of the samples tested range from 0.3 to 8 μ and average about 3 μ. For the three samples on which both types of data are available, the Atterberg limits vary inversely with the logarithm of the median diameter, but three samples are, of course, too few to permit interpretation with assurance. The relationship of the Atterberg limits to grain size distribution is under investigation at the present time.

HISTORY OF LAKE AGASSIZ

Lake Agassiz occupied a large area in North Dakota, Minnesota, and adjacent parts of Canada at the close of the last (Wisconsin) glaciation. At its maximum extent, water covered nearly as much area as that of all the present Great Lakes combined. Glacial ice formed the northeast boundary of the lake, damming the water in the valley now occupied by the Red River of the North. The nature of this damming, whether by retreating ice, as advocated by Upham (1895) and Leverett (1932), or by advancing ice blocking the northeastward drainage, as advocated by Tyrell (1896) and Johnston (1916), is a controversial topic. Discussion of this problem is beyond the scope of the present paper. It suffices to state here that, in general, the lake became successively shallower and occupied smaller areas with the passage of time.

Old beaches, indicated by present-day gravel ridges, mark the position of the southern border of the lake at its various stages. Fifty such beaches have been studied and described in considerable detail by Johnston (1946, fig. 2). The index map (fig. 1) shows three of the most prominent beaches; the Herman beach is the highest and records the maximum extent of the lake, the Campbell beach is interpreted by Johnston (1916) as the first beach after the lake had been drained temporarily through some eastern outlet, and the Burnside beach is the first one lower than Grand Forks.

The sediments discussed in the present paper were deposited during the time when the lake stood at various levels between the highest Herman

beach and the Burnside beach. According to Johnston and Antevs (Johnston, 1946, pp. 17–18), these beaches were formed about 23,000 and 9,000 years ago, respectively; hence the sediments under consideration were deposited over a period of approximately 14,000 years.

According to Laird (1944, pp. 22–23), the unconformity at the top of unit 4 of the present paper probably corresponds to that observed in the Lake of the Woods region by Johnston and interpreted by him as indicating the period of eastward drainage of Lake Agassiz just prior to the formation of the Campbell beach (Johnston, 1946, p. 3). Thus the shore lines at the time of deposition of units 1 through 4 stood between the highest Herman beach and the Campbell beach, and the shore line during deposition of unit 5 was at the Campbell beach or between the Campbell beach and the Burnside beach.

The base of the 53 feet of clay comprising units 1 through 4 at Grand Forks is at an elevation of 434 feet below the highest Herman beach and 214 feet below the Campbell beach, the elevations of these beaches being taken at points in the direction of the isobase from Grand Forks, so that differences in elevation are between points of equal uplift.[4] Therefore, the depth of the water during deposition of units 1 through 4 varied from about 434 to 214 feet at Grand Forks, except during formation of the drying surface at the top of unit 3. Similarly, the base of the 20 feet of silt and clay constituting unit 5 at Grand Forks is 214 feet below the Campbell beach, indicating that the sediments were deposited when the lake was 214 feet or less in depth. The nature of these sediments suggests a depth considerably less than 200 feet. The corresponding depths at Fargo are about 310–125 feet for units 1 through 4 and less than 125 feet for unit 5; at Crookston about 430–185 feet for units 1 through 4 and less than 185 feet for unit 5. These data are summarized in table 7, which includes the various beach elevations; the elevations are interpolated from maps by Leverett (1932) and by Johnston (1946), and the trends of isobases (shown in fig. 1) are from Johnston (1946).

Results of the present investigation necessitate modification of the previous concepts of the history of Lake Agassiz to account for the development of the drying surface at the top of unit 3. The surface drying indicates that the lake bed, at least over the area considered in this paper, was completely drained prior to the drainage period recognized by previous workers. The lithologic similarity of the clay above and below the old drying surface and the lack of noticeable disconformity suggest that this additional drying period was of short duration—i.e., that the opening of the drainage outlet was followed almost immediately (perhaps even within a few years) by closure of the same outlet.[5] In this case the most likely agent of such successive opening and closing of an outlet is, of course, glacial ice, and it is probable that some divide to the northeast was uncovered by retreating ice and covered again by readvancing ice, thus causing the temporary drainage. If this is true, a stage of ice advance instead of a period of continuous ice re-

[4] For a recent discussion of the tilting of beaches due to recovery of the earth's crust from the load of glacial ice and of the delineation of isobases, or lines of equal uplift, see Johnston, 1946.

[5] On the basis of measurements of varved clays, Rittenhouse (1934) has recognized a drainage of 24 years' duration in northern Ontario, and much shorter periods of drainage have been recognized by Antevs in eastern North America.

treat is indicated, and the portion of the history of Lake Agassiz that occurred during ice retreat, as advocated by Upham (1895), must be restricted even more than is necessitated by the work of Johnston (1916). It should be pointed out, however, that the opening and closing of an outlet by ice retreat and readvance of the most far-reaching effect is that the drainage was of short duration but that subsequent conditions of deposition were similar to those prior to the drainage. And this indicates that the water rose rapidly after the drainage until it reached a depth commensurate with that prior to the drainage, a situation that is entirely possible with ice damming. Hence, although there was

TABLE 7

DEPTHS OF LAKE AGASSIZ DURING DEPOSITION OF STRATIGRAPHIC UNITS 1–4 AND OF STRATIGRAPHIC UNIT 5*

	Grand Forks (Feet)	Crookston (Feet)	Fargo (Feet)
Measured data:			
A. Elevation of highest Herman beach	1,187	1,195	1,113
B. Elevation of Campbell beach	1,020	1,021	998
C. Elevation of base of unit 1	753	766	802
D. Elevation of base of unit 5	806	835	872
E. Elevation of present surface	826	853	887
Derived data:			
1. Depth of water during deposition of units 1 through 4			
Maximum (=A−C)	434	429	311
Minimum (=B−D)	214	186	126
2. Depth of water during deposition of unit 5			
Maximum (=B−D)	214	186	126
Minimum (indeterminate, but must be greater than 0)	0	0	0

* As determined from differences in elevation between the bases of the units and their corresponding beaches, measured along the isobases through the three localities.

recorded in the unconformity above unit 4; the break at the old drying surface above unit 3 is of relatively minor importance, except for the time of its occurrence. It is noteworthy also that the postulated eastward drainage is in complete accordance with the physical features of the region and is known to have occurred at several times slightly later in the history of the lake (see Johnston, 1946; Rittenhouse, 1934).

The fact that the old drying surface occurs between clays of very similar character and is not evidenced by marked disconformity suggests not only definitely a time of no water at all and a time of only shallow water, as substantiated by the presence of shells and silt at Fargo, a 125– to 430–foot depth of the lake probably persisted throughout the deposition of units 1 through 4, with but short interruption.

The depositional history of the sediments studied in the present investigation may be summarized as follows: (1) deposition of the clays of units 1 through 4 in a portion of Lake Agassiz that was from about 430 to 125 feet deep, except for a short period of complete drainage and surface drying; (2) major drainage

of the lake for a period long enough to include significant erosion; and (3) deposition in shallower water of the silt and clay of unit 5. The corresponding history of ice movement is interpreted as (1) general retreat of ice to the northeast until an outlet was uncovered, followed almost immediately by closure of the outlet by readvancing ice; (2) retreat of the ice farther to the northeast, leaving an outlet open in that direction for a relatively long time; and (3) readvance of the ice, causing ponding of the water into a lake of shallower depth than the earlier stages.

CONCLUDING REMARKS

The primary purpose of the present investigation is to show the value of the use of soil mechanics data in geology. In the particular case studied, the data have provided means of (1) correlating lithologic units not easily defined by ordinary geologic methods and (2) locating an old drying surface that indicates a hiatus not otherwise apparent. Study of the cores has also yielded information of a type more commonly used in geology, namely, lithology and elevations of the tops of stratigraphic units. The interpretation of the history of Lake Agassiz on the basis of these soil mechanics and geologic data is given not only for its own value but as an illustration of the efficacy of the combined use of data from the two fields.

Similar soil mechanics tests have been made in foundation engineering studies for numerous construction projects. In many such instances the data would no doubt be available for geologic study. The opportunity for joint research on problems similar to the one discussed in this paper is particularly good in universities having both geologic and soil mechanics departments. It is the opinion of the authors that such joint study would yield considerable information of value to both the geologist and the soil engineer.

Among problems having the closest relation to the present study, the following would perhaps be the most fruitful fields for future work: (1) study of the interrelationships of particle size distribution, mineralogic composition, and Atterberg limits of clays; (2) study of the areal extent of the various stratigraphic units of Lake Agassiz sediments and their lake beaches, with the goal of constructing detailed maps showing the extent of each unit; (3) study of the areal variation of geologic and soil mechanics properties of the sediments of each unit over its entire area of occurrence; (4) geologic field study of the nature of the old drying surface indicated by soil mechanics properties; and (5) integration of the above information with the detailed history of Lake Agassiz. Various phases of these problems are now being investigated by the authors.

ACKNOWLEDGMENTS.—Soil samples, boring records, and test data from Grand Forks and Fargo, North Dakota, were made available by the Northern States Power Company, Mr. Hibbert M. Hill, hydraulic engineer. Samples and test data from Crookston, Minnesota, were furnished by the Otter Tail Power Company, Mr. Cyrus G. Wright, vice-president, and Mr. G. W. Welch, chief engineer. The test borings and undisturbed sample borings were made by the Gow Division of the Raymond Concrete Pile Company. Test-pit samples were obtained by the power-company forces under the supervision of Mr. Norris Fitch at Grand Forks, and of Dr. J. O. Osterberg, Northwestern University, and Mr. G. W. Welch at Crookston. Laboratory soil tests were performed by Messrs. John L. McRae, T. W. Van Zelst, and others of the staff of the Northwestern University Soil Mechanics Laboratory, under Dr. Osterberg's supervision. The soil tests were designed and performed to obtain a complete en-

gineering picture of the underground conditions for foundation analysis. The writers wish to express appreciation to all concerned for their contributions to the data used herein.

The writers are also indebted to Dr. W. C. Krumbein, Northwestern University, for helpful suggestions and for critical reading of the manuscript.

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THE STABILITY FIELD OF SAPPHIRINE IN THE SYSTEM MgO-Al_2O_3-SiO_2[1]

M. L. KEITH[2] AND J. F. SCHAIRER[3]

ABSTRACT

A detailed study by the method of quenching of ten closely spaced compositions in a small portion of the ternary system MgO-Al_2O_3-SiO_2 has delineated the tiny field of sapphirine and located the temperatures and compositions of three ternary reaction points. Sapphirine crystals are stable in equilibrium with a liquid in the ternary system only over the narrow range of temperature between 1,482° and 1,453° C.

INTRODUCTION

W. R. Foster (1950*a*, *b*) published results of an investigation of sintered preparations in the high-alumina part of the system MgO-Al_2O_3-SiO_2. He obtained crystalline sapphirine essentially identical in X-ray properties with natural sapphirine, deduced the most probable composition for synthetic sapphirine as $4MgO \cdot 5Al_2O_3 \cdot 2SiO_2$, and concluded that it must have a primary stability field on the liquidus surface of MgO-Al_2O_3-SiO_2.

At the time Foster's paper appeared, experimental studies were in progress at the Geophysical Laboratory on two different quaternary systems[4] which have the system MgO-Al_2O_3-SiO_2 as one of the limiting ternary systems. Therefore, it seemed desirable to check Foster's prediction that sapphirine should have a primary stability field and to determine the limits of sapphirine stability. A discussion of the relations of sapphirine to other phases in the two quaternary systems under investigation will be published in the near future. The present report refers only to results obtained in the ternary system MgO-Al_2O_3-SiO_2.

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[4] FeO-MgO-Al_2O_3-SiO_2 (Keith); K_2O-MgO-Al_2O_3-SiO_2 (Schairer).

EXPERIMENTAL PROCEDURE

Ten compositions were prepared for study by the method of quenching (Shepherd, Rankin, and Wright, 1909). These compositions were selected to cover that small portion of the system MgO-Al_2O_3-SiO_2 where a primary field of sapphirine would lie. Melts were prepared in platinum crucibles from purified quartz (0.03 per cent residue after treatment with sulfuric and hydrofluoric acids), C.P. MgO (J. T. Baker Co., lot No. 82735), and tabular alumina T61 (free from iron) obtained from the Aluminum Company of America. Five fusions at 1,600° C., with intermediate crushing in a steel mortar and removal of steel chips magnetically, were necessary to obtain a homogeneous glass of the desired composition. All these compositions crystallize readily, and small quenching charges of crushed glass were used and found to crystallize in the few minutes required to bring them to the temperature of the quenching run. In this way no large crystals were formed, and it was possible to get equilibrium between crystals and liquid in a few hours (5 hours or less). The Pt-Pt90Rh10 thermoelement was calibrated before and after each quenching run with pure diopside ($CaO \cdot MgO \cdot 2SiO_2$) the melting point of which is defined as 1,391.5° C.

RESULTS OF QUENCHING EXPERIMENTS

The results of quenching experiments are given in table 1. From these results the temperatures and compositions of the three ternary invariant points with sapphirine as one of the crystalline phases can be located and are given in table 2.

From an examination of the quenching data given in table 1 it may be seen that the composition No. 59S gives the best data for the location of the temperature and composition of the ternary invariant point X (fig. 1) and the composition 58S gives the best value for W (fig. 1). The point N (fig. 1) may be located from the data on Nos. 57S, 55S, 51S, and 53S.

Figure 1 is the revised diagram for MgO-Al_2O_3-SiO_2. It shows the small stability field (XNW) of sapphirine. This diagram was prepared for use by Schairer in a paper now in preparation on a portion of the quaternary system K_2O-MgO-Al_2O_3-SiO_2, and for this use Al_2O_3 was placed at the upper apex of the equilateral triangle, with MgO as the lower

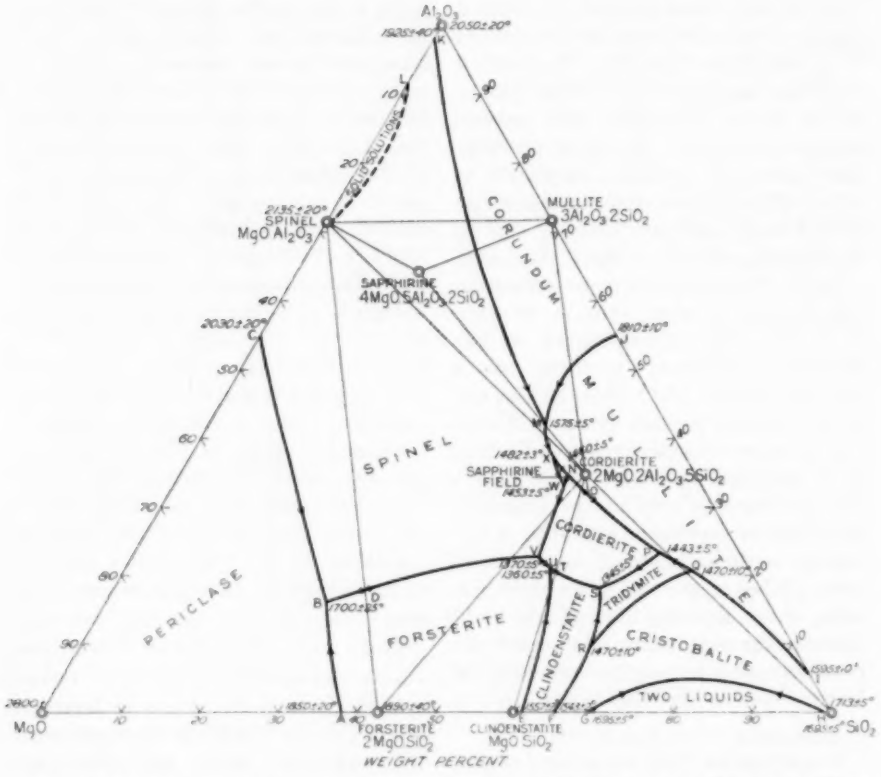

FIG. 1.—Revised equilibrium diagram for the system MgO-Al_2O_3-SiO_2 based on the original work of Rankin and Merwin (1918) as modified by Bowen and Greig (1924), Greig (1927), Schairer (1942), Foster (1950*a*, *b*), and Keith and Schairer (this paper). Recent studies by Foster (1951), published while this paper was in galley proof, indicate that the field labeled "clinoenstatite" should read "protoenstatite."

TABLE 1

RESULTS OF QUENCHING EXPERIMENTS

MELT NO.	COMPOSITION (WEIGHT PER CENT)			TEMP. (° C.)	TIME (HOURS)	RESULTS OF MICROSCOPIC EXAMINATION
	MgO	Al_2O_3	SiO_2			
57S.....	16	38	46	1,510	4	All glass
				1,505	4	Small amount mullite in glass
				1,485	5	Lots mullite in glass
				1,480	5	Lots mullite and rare sapphirine in glass
				1,480	19	Lots mullite and rare sapphirine in glass
				1,465	19	Lots mullite and small amount sapphirine in glass
				1,460	18	Lots mullite, small amount cordierite, small amount sapphirine in glass
				1,455	18	Lots cordierite, rare mullite, and rare sapphirine in glass
55S.....	16	37	47	1,515	2	All glass
				1,510	2	Rare mullite in glass
				1,480	19	Lots mullite in glass
				1,475	20	Lots mullite and small amount sapphirine in glass
				1,465	5	Lots mullite and moderate amount sapphirine in glass
				1,460	23	Moderate amounts of mullite and sapphirine, and rare cordierite in glass
				1,455	19	Lots cordierite and moderate amount sapphirine in glass
51S.....	16	36	48	1,505	2	All glass
				1,500	4	Rare mullite in glass
				1,470	23	Moderate amount mullite in glass
				1,465	24	Lots mullite and rare sapphirine in glass
				1,460	20	Lots mullite, small amount sapphirine, and rare cordierite in glass
				1,455	19	Lots cordierite, moderate amount sapphirine, and rare mullite in glass
53S.....	16	35	49	1,490	4	All glass
				1,485	4	Rare mullite in glass
				1,475	6	Lots mullite in glass
				1,470	3	Lots mullite and very rare sapphirine in glass
				1,465	18	Lots mullite and small amount sapphirine in glass
				1,460	18	Moderate amount mullite, small amount cordierite, and small amount sapphirine in glass
				1,455	19	Lots cordierite, rare mullite, rare sapphirine, and rare spinel in glass
59S.....	17	37	46	1,505	2	All glass
				1,500	2	Rare spinel in glass
				1,490	4	Small amount spinel in glass
				1,485	5	Moderate amount spinel and very rare mullite in glass
				1,480	5	Moderate amount spinel, rare mullite, and rare sapphirine in glass
				1,480	18	Moderate amount spinel, rare mullite, and rare sapphirine in glass
				1,475	20	Moderate amount sapphirine in glass
				1,460	23	Lots sapphirine in glass
				1,455	5	Lots sapphirine and rare spinel in glass
				1,450	20	Moderate amounts cordierite and spinel in glass
50K.....	17	36	47	1,490	6	All glass
				1,485	6	Rare spinel in glass
				1,475	6	Moderate amount spinel in glass
				1,470	19	Lots spinel and rare sapphirine in glass
				1,460	22	Moderate amount sapphirine in glass
				1,455	5	Moderate amount sapphirine and rare cordierite in glass
				1,450	7	Lots cordierite, moderate amount spinel, and rare sapphirine in glass
				1,445	24	Lots cordierite and moderate amount spinel in glass

TABLE 1—*Continued*

Melt No.	Composition (Weight Per Cent)			Temp. (° C.)	Time (Hours)	Results of Microscopic Examination
	MgO	Al_2O_3	SiO_2			
54S	18	34	48	1,480	2	All glass
				1,475	3	Rare spinel in glass
				1,460	23	Small amount spinel in glass
				1,455	5	Small amount spinel and rare cordierite in glass
52S	17	35	48	1,475	6	All glass
				1,470	23	Rare sapphirine in glass
				1,460	20	Small amount sapphirine in glass
				1,455	19	Moderate amounts cordierite and spinel, and rare sapphirine in glass
				1,450	3	Lots cordierite and moderate amounts spinel in glass
				1,450	18	Lots cordierite and moderate amounts spinel in glass
58S	17	34	49	1,470	5	All glass
				1,465	18	Very rare sapphirine in glass
				1,460	18	Small amount sapphirine and very rare cordierite in glass
				1,455	18	Lots cordierite and small amounts of spinel and sapphirine in glass
				1,450	5	Lots cordierite and moderate amount spinel in glass
				1,450	18	Lots cordierite and moderate amount spinel in glass
56S	18	33	49	1,460	18	All glass
				1,455	5	Rare cordierite and spinel in glass
				1,455	19	Rare cordierite and spinel in glass
				1,450	18	Small amounts cordierite and spinel in glass

left apex and SiO_2 as the lower right apex. Attention is called to the value 1,443° ± 5° C. at *P* (fig. 1), the ternary invariant point cordierite+mullite+tridymite+liquid. This differs from the value 1,425° ± 5° C. given by Rankin and Merwin (1918), and the data on which the new value is based will be given by Schairer in his paper on K_2O-MgO-Al_2O_3-SiO_2.

DISCUSSION OF THE RESULTS

From an examination of figure 1 it may be seen that all compositions within the area corundum-mullite-spinel begin to melt on heating or, conversely, become completely crystalline on cooling only at 1,575° ± 5° C., the temperature of the ternary invariant point *M* (fig. 1). Compositions within the area spinel-mullite-sapphirine begin to melt at *X* at 1,482° ± 3° C.; those within the area mullite-sapphirine-cordierite begin to melt at *N* at 1,460° ± 5° C.; those within the area sapphirine-cordierite-spinel begin to melt at *W* at 1,453° ± 5° C.; while those within the area forsterite-spinel-cordierite begin to melt at *V* at 1,370° ± 5° C.

It should be noted that the data given in table 2 and presented graphically in figure 1 indicate a narrower range of temperature for sapphirine stability than that suggested by Foster (1950*a*, *b*). However, our results confirm his deductions in every important respect except that we place the temperature of the reaction point spinel+cordierite=sapphirine+liquid at 1,453° ± 5° C. rather than at 1,410° C. Foster's evidence for placing the temperature of that point as low as 1,410° C. depended upon his observation of "shrinkage and partial melting" in spinel-cordierite mixtures (Foster, 1950*b*, pp. 76, 81). Some of the possible explanations for Foster's obser-

vation of the presence of liquid in spinel-cordierite mixtures below 1,450° C. might be: (*a*) there might be some excess Al_2O_3 over the 1:1::MgO:Al_2O_3 ratio in the spinel crystals in such mixtures; (*b*) if these mixtures were even slightly off-composition in the direction of excess MgO, they would lie in the triangle spinel-cordierite-fosterite and show the presence of some liquid down to 1,370° ± 5° C., the temperature of *V* (fig. 1); (*c*) the presence of small quantities of impurities, such as alkalies, in the ingredients used to prepare the mixtures would result in the presence of appreciable amounts of liquid, but no crystalline sapphirine, however, at temperatures below 1,453° C.

TABLE 2

TERNARY REACTION POINTS INVOLVING SAPPHIRINE

LETTER (FIG. 1)	COMPOSITION (WEIGHT PER CENT)			TEMP. (° C.)	PHASES PRESENT
	MgO	Al_2O_3	SiO_2		
X	16.9	36.8	46.3	1,482±3	Spinel, mullite, sapphirine, and liquid
N	16.3	34.4	49.3	1,460±5	Mullite, sapphirine, cordierite, and liquid
W	17.4	33.5	49.1	1,453±5	Sapphirine, spinel, cordierite, and liquid

PROPERTIES OF SYNTHETIC SAPPHIRINE

A sample of synthetic sapphirine crystals mixed with some glass was obtained by centrifuging a crushed charge from one of the quenching experiments. The crystals gave an X-ray pattern identical with that given by Foster (1950*b*, p. 76). They are monoclinic, tabular ⊥ *Y*, with:

$Z \wedge c = 15°$;
Mean index of refraction 1.705 ± 0.003;
$Ng - Np$ probably less than 0.003;
(−) $2V = 55 \pm 5°$ (universal stage).

It was found difficult to measure the indices of refraction more precisely because of low-index glass attached to the crystals.

The sapphirine crystals which are present in the quenching charge are easily distinguished from spinel. They occur as elongated and usually faceted monoclinic crystals with a low but appreciable birefringence, whereas spinel always occurs in these melts in small but perfect isotropic octahedrons. In one preparation one twinned sapphirine crystal was observed but was lost before any observations on the twinning could be made.

COMPOSITION OF NATURAL AND SYNTHETIC SAPPHIRINES

The significance of the data for synthetic sapphirine in regard to the occurrence of natural sapphirine will be discussed as part of forthcoming reports. The formula which Foster deduced is the most probable formula for synthetic sapphirine. Some work has been done on the crystal structure of sapphirine, and more may be necessary. Gossner and Mussgnug (1928) used the formula $2MgO \cdot 2Al_2O_3 \cdot SiO_2$, and their X-ray data and the phase equilibrium data must be correlated.

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GEOLOGICAL NOTES

INTERRELATIONS OF GEOMORPHOLOGY, GLACIAL GEOLOGY, AND PLEISTOCENE GEOLOGY[1]

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The close relationships between glacial geology, Pleistocene geology, and geomorphology are generally recognized and are indicated by the fact that many geologists work in all three fields. In many situations, methods and purposes are so nearly identical that distinctions become meaningless. Yet there are essential differences. Pleistocene research is broader in scope than either glacial geology or geomorphology and embraces many fields of study. It includes biogeography, climatology, archeology, and pedology, which are not strictly geology, and it includes also such geological subjects as paleontology, stratigraphy, and marine geology, which normally lie outside the scope of both geomorphology and glacial geology. Glacial geology shares with geomorphology its interest in glacial land forms but diverges from it in its concern with glacial sediments and pre-Pleistocene glaciations.

The principal ties between geomorphology and the combined fields of glacial geology and Pleistocene geology are: (1) the mutual concern with glacial landscapes; (2) the important influence of topography on the growth and development of both mountain and continental glaciers; (3) the eustatic changes in sea-level which accompanied Pleistocene glaciation and are reflected in a variety of coastal and marine features; (4) the diastrophic and volcanic events of the Pleistocene, which are intimately related to both landscape development and glaciation; and (5) a common interest in Pleistocene chronology based on correlation and dating by diastrophic, volcanic, glacial, and climatic events.

This latter association with earth history, which strikes close to the common ground of all geology, was pointed out by Bryan (1941, p. 5):

> If land forms were solely the result of processes now current there would be no excuse for the separation of the study of land forms as a field of effort distinct from Dynamic Geology. The essential and critical difference is the recognition of land forms produced by processes no longer in action. Thus, in its essence and in its methodology, physiography is historical. Thereby it is a part of Historical Geology, although the approach is by a method quite different from that commonly used.

Accepting this point of view, the central thesis of this discussion may be stated as three propositions: (1) that most landscapes are complex rather than simple and many include a predominance of relict features which often are not recognized as such; (2) that these landscape features are largely of Pleistocene age; and (3) that the geomorphic events recorded by the complex landscape are best correlated on the basis of climatic fluctuations of the Pleistocene. In this light a central aspect of geomorphology is its application to the task of deciphering Pleistocene history.

Hence it is important to consider the methods and scope of geomorphic history, the soundness and feasibility of climatic correlation, and the implications in terms of traditional geomorphology.

The view that geomorphology is a phase of earth history certainly is not new, and the interpretation of geomorphic history is a

[1] Manuscript received October 22, 1951.

goal actively pursued by a large group of geomorphologists. It is not, however, the geomorphology presented formally in textbooks and classrooms. As an indirect and unintentional result of the teachings of Davis, emphasis is placed on present processes, or a single dominant process of the past, and on the normal fluvial cycle for humid-temperate regions. The notion that land forms are largely the products of present conditions and that the cycle is the end of geomorphic interpretation is a misconception commonly encountered both in the classroom and outside. Actually, the normal form and the simple landscape are rarities. Most landscapes are complex and are the result of multiple cycles and epicycles, the records of which are often masked by the impress of later events and must be interpreted from relict features. The simple case of the normal cycle, instead of pointing up exceptions in the complex landscape, has, in its facility of application, often been an end in itself. The result has been simply classification of stage and physiographic description. A complete understanding of landscape, as was recognized by Davis (1915, pp. 76–77), requires more than an analysis of form. It demands a consideration of history as well as the traditional triumvirate of structure, process, and stage.

The historical approach broadens the field of geomorphology to include the application of the geological methods of sedimentology and stratigraphy and attention to such related fields as soil science and climatology.

It also imposes the responsibility of developing a body of principles by which geomorphic history can be interpreted. These principles as such are seldom considered and are yet to be defined and organized. As an initial step, it may be desirable to recognize four general types of landscapes, suggested by Bryan's (1943, pp. 472–473) classification of soils: (1) simple, monocyclic landscapes which can be fitted to the idealized cycle; (2) complex, multicyclic landscapes which require historical interpretation; (3) compound landscapes which are land surfaces mantled by a cover of younger sediments but retaining distinctive original characters; and (4) exhumed landscapes which are due to stripping of sediments above an unconformity. It also may be desirable, in a search for new approaches, to apply familiar stratigraphic concepts leading to consideration of such topics as: (1) the nature and significance of geomorphic unconformities; (2) criteria for determining relative age of cyclical surfaces; (3) bases for correlation of erosional and depositional surfaces; and (4) the concept of facies applied to landscapes. As a basis for such consideration it will be necessary to secure additional information on the profiles of weathering on erosional and depositional surfaces, the areal distribution of surfaces, the succession of related deposits, and the nature of the deformation or climatic change responsible for rejuvenation.

The historical approach emphasizes the need for recognition of relict or "fossil" features inherited from past conditions and especially those relict features which are subordinate elements in the over-all landscape. In some areas they are impressed on landscapes which are largely the result of present processes; in other areas they appear in landscapes, such as youthful drift plains, which themselves record a dominant process of the past.

Many of the fossil features in present landscapes are the result of periglacial epicycles. They include: slopes molded by periglacial mass movements, block fields, solifluction deposits, earth mounds and ridges, stabilized debris accumulations in mountains, dry valleys, asymmetric valley slopes, and ancient dune complexes (Smith, 1949*a*). They are known to extend across the interior of the continent and are exemplified by: rubble-clad slopes and solifluction deposits in the Appalachians (King, 1950, p. 37; Peltier, 1949, pp. 48–74); block fields in the Driftless Area of Wisconsin (Smith, 1949*b*); similar features in the St. François Mountains of Missouri (Peltier, 1950, pp. 231–232); minor ridges on the floor of the

Lake Agassiz Basin (Horberg, 1949); dune complexes scattered widely over the Great Plains (Melton, 1950; Smith, 1940, pp. 153–168); ancient landslides, protalus ramparts, structure soils, and talus accumulations in the western mountains; and possibly natural mounds in the Puget Sound lowland. In certain areas these features may be dominant elements of the landscape, but in most areas they are minor. From the historical point of view, however, all are significant. They are significant also to dynamic geology, in that relict features, such as slope deposits, need to be recognized as such and not confused with the recent products of present processes. The fact that many of these features are periglacial points to the need of considering the periglacial cycle as it now operates in polar regions in contrast to the normal fluvial cycle of Davis. In this, geomorphologists and Pleistocene geologists certainly have much in common.

Aside from periglacial and glacial features, the fluctuating climatic events of the Pleistocene penetrate into a variety of landscapes over the entire world. Eustatic changes of sea-level are reflected in marine and fluvial terraces, stream profiles, valley fills, the configuration of shorelines, coral reefs, and possibly submarine canyons. Climatic fluctuations are revealed by stream terraces, ancient dune fields, pluvial-lake features, and complex soils.

In addition to the climatically controlled land forms, there are the large-scale features due to Pleistocene diastrophism and resultant erosion. It is to be noted (1) that the record of Pleistocene diastrophism is based in large part on geomorphic, as well as structural and stratigraphic evidence; (2) that diastrophism was world wide and continued over a large part of Pleistocene time; and (3) that it is both a complicating and an assisting factor in Pleistocene chronology. It is strikingly revealed in the Rocky Mountains by the profound canyon-cutting, in many areas amounting to over 1,000 feet, which intervened between the pre-Wisconsin (Kansan) and later Wisconsin glaciations.

Because the details of landscape are largely Pleistocene in age, it appears that the climatic fluctuations of the Pleistocene provide the best hope for geomorphic correlation. These fluctuations are well known from the glacial and postglacial record, and their major pulsations appear to be firmly established. The sequence of major events would include: four principal stages of glaciation, separated by interglacials; three or four substages of the last glacial; and a postglacial stage in which early arctic and subarctic conditions were followed by a climatic optimum and finally by the "Little Ice Age." Uncertainties still exist as to the number of substages in the earlier stages of glaciation, but there is hope that these may be resolved with detailed studies, especially of the subsurface record. The data available from both hemispheres on lowering of snow lines during the last glacial stage, the behavior of glaciers in historic time, and submarine cores indicate that climatic fluctuations were synchronous over the entire earth and theoretically are feasible as bases for correlation.

In application the methods of correlation will vary greatly. Within the glaciated and former periglacial areas glacial deposits and periglacial features offer a direct method of correlation. Outside these areas eustatic features are world wide in extent and become usable in coastal areas where diastrophic movements can be evaluated. In nonglaciated areas toward the interiors of the continents the influence of climatic changes on processes becomes the only method available. The evidence at hand seems to indicate that pluvial climates coincided with glacial stages and that climatic change in many areas was able to bring about changes in stream regimen, eolian activity, mass movements, soil development, and other processes, Further studies in these areas will be needed before a consistent Pleistocene chronology can be realized.

The earth-history view of geomorphology contends that geomorphologists, glacial

geologists, and Pleistocene geologists in one essential aspect, that of Pleistocene physical history, are engaged in a common effort. It implies a break with traditional geomorphology, which, as it is formally presented, is too often an advanced course in geologic processes. It requires a broader scope of field investigation, in which attention is given to sediments, stratigraphic relations, internal structures, and soils. It necessitates that interpretations be made in the light of climatic and diastrophic history. It emphasizes the complex landscape and all related deposits. It points to the immediate goals of discovering critical sections and significant field relations and of recognizing in them the possibilities for climatic interpretation and correlation. It indicates the need for a body of principles which will provide a basis for the full understanding of landscape in terms of earth history.

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A SOLID SOLUTION BETWEEN CALCITE AND DOLOMITE[1]

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Combined X-ray and chemical studies of living and fossil organisms have demonstrated a partial solid solution between the minerals calcite, $CaCO_3$, and dolomite, $CaMg(CO_3)_2$. This series, deposited by living organisms, is apparently the only natural occurrence, for inorganically precipitated calcites almost invariably show less than 2 per cent $MgCO_3$. Typical analyses of these inorganic minerals are given by Kulp, Kent, and Kerr (1951). Clarke and Wheeler (1922) and others have shown clearly that many organisms deposit relatively high concentrations of magnesium in their hard parts. This element was generally believed to be present as dolomite, although the latter was never found by optical or X-ray examination.

The relation between weight per cent $MgCO_3$, chemically determined, and the *d*-value parallel to the major cleavage (100) is shown in figure 1. These data show that a single phase is present, containing from 0 to 29 per cent $MgCO_3$, but above 16 per cent and below 2 per cent there are devia-

[1] Manuscript received November 2, 1951.

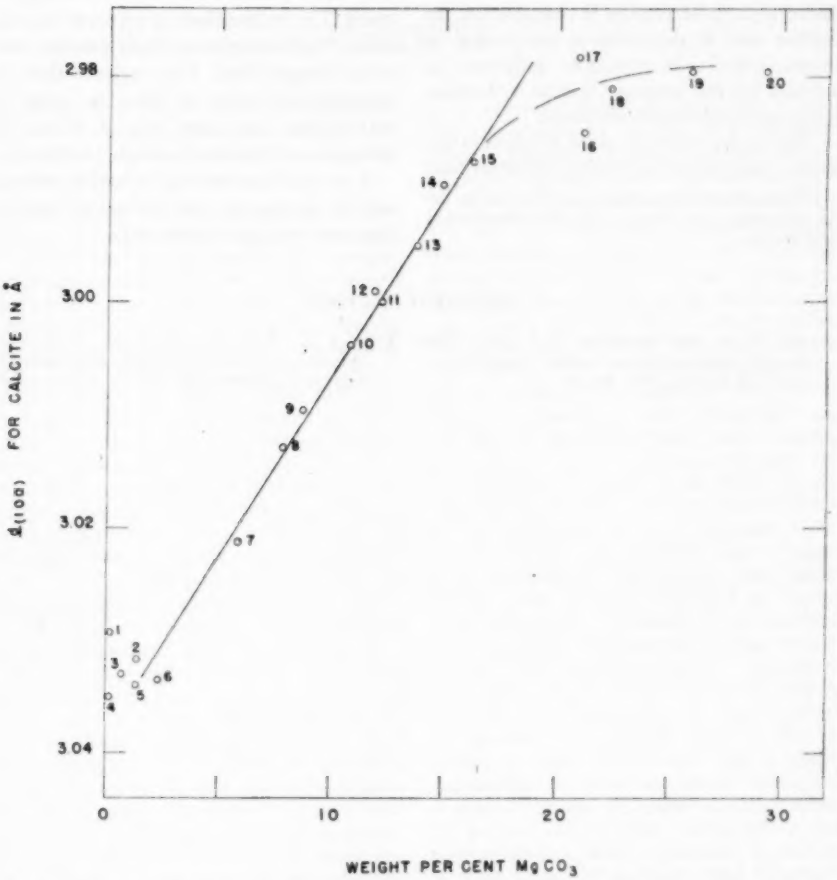

FIG. 1.—The relation between magnesium content and interplanar spacing. The analyzed specimens in the graph are (*1*) pelecypod (Bermuda); (*2*) pelecypod (Alaska); (*3*) pelecypod (Bermuda); (*4*) Iceland spar (unknown); (*5*) pelecypod (Bermuda); (*6*) barnacle (Japan); (*7*) sponge (California); (*8*) echinoid (California); (*9*) echinoid spines (Bermuda); (*10*) echinoid (Bermuda); (*11*) echinoid (Guam); (*12*) Foraminifera (Bermuda); (*13*) coral (Bermuda); (*14*) algae (Bermuda); (*15*) starfish (Florida); (*16*) algae (Guam); (*17*) algae (Palau); (*18*) algae (Bermuda); (19) algae (Florida); and (*20*) algae (Florida).

tions from Vegard's law.[2] In both areas of deviation, less change in spacing is produced by a given change in composition. In neither area of deviation is the change in trace-element concentration sufficient to account for the anomaly (Heinz A. Lowenstam, personal communication).

The calcite-dolomite solid solution is unstable. Analyses of fossil materials show that the magnesium content falls to 1 or 2 per cent often within a few tens of millions of years. The magnesium is replaced by calcium. The forms richest in magnesium seem to be changed first. This suggests that the solid-solution series is unstable under all near-surface conditions, except within the biological environment which produced it.

A further discussion of this solid solution and its geological and biological implications will be published shortly.

[2] Vegard's law states that in a solid solution the cell dimensions vary linearly with the concentration of the solute.

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REVIEWS

Economic Mineral Deposits. By A. M. BATEMAN. 2d ed. New York: John Wiley & Sons, Inc.; London: Chapman & Hall, Ltd., 1950. Pp. xi+916; figs. 302; tables 25. $7.50.

This new edition of Bateman's *Economic Mineral Deposits* is highly welcome because the additions of the last eight years to the literature of economic geology badly need to be integrated into a systematic whole. The first edition of this book (reviewed in this *Journal* by E. S. Bastin, 1943) represented in many ways a marked advance over previous books in the field. The book was well organized, clearly written, and, considering the conditions under which it was written and published, quite free of errors. Its main defects were noted by Bastin in his review. The new edition is superior to its predecessor in organization, content, and typographic accuracy.

The book retains the sound outline of the first edition and is divided into the same three parts: I: "Principles and Processes"; II: "Metallic Mineral Deposits"; and III: "Nonmetallic Mineral Deposits." The details of textual arrangement in the new edition have been changed somewhat over the old, a good many pages have been rewritten with some minor additions of new material, but few entirely new sections have been added. Some of the rewriting has been to clarify or make more logical portions of the first edition, and some has been to present the sense of the important papers of the last ten years. Many of the important papers of the decade just past, however, are noticed only as annotated references in the bibliographies at the ends of the various chapters and chapter subdivisions. It is to be regretted that this procedure has been followed, because it requires that the student users of the book, at least, must first read a given chapter in Bateman's book, then read these cited papers, and, finally, in effect rewrite Bateman's text to achieve a proper balance among the ideas presented. Surely this is the task of the author and not of the student. For example, few students have the training and experience necessary to evaluate the work of Smith referred to on page 67 in the light of the discussion of Fenner's ideas presented in chapter iv.

In a review of a second edition, there is space only for comments on the most important changes appearing in the revised text; what is largely unchanged need be discussed only if it appears that it should have been changed and was not. The most striking changes, primarily rearrangements combined with some new connective material, occur in chapter v. In the first edition Bateman put forward a classification of ore deposits which did not contain the phrase "hydrothermal processes" at all. He discussed the processes which produced such deposits under the headings of "replacement" and "cavity filling" in subsections 4 and 5 of chapter v and classified the deposits formed by these processes as "replacement," "replacement-filling," and "cavity filling" in Part II. As Bastin pointed out, this constituted a serious defect in Bateman's classification, but it has now been partly remedied by grouping and discussing, in chapter v, the processes of "replacement" and "cavity filling" under the general subject of "hydrothermal processes." It seems to me, however, unfortunate that the descriptive method of subdividing "hydrothermal processes" was used in the second edition, particularly because Bateman subdivided a category of equal theoretical importance—magmatic concentration—on genetic grounds. Admittedly, the Lindgren classification needs revision, but its subdivision of deposits formed by hydrothermal processes by geochemical intensity (temperature and pressure ranges during deposition) appears to be a much sounder approach than Bateman's (which puts Bisbee, Ariz., and Mascot, Tenn., under the same subheading), as well as much more easily further subdivided by a genetic scheme.

Now that "hydrothermal processes" have been introduced into the book, it is difficult to see why the ore deposits of contact metamorphic, or, as Bateman now prefers, contact metasomatic, origin should not be included under this heading, for water (whether as a gas or as a liquid) very probably is the transporting medium for the ore-forming ions in such ore bodies. Apparently, Bateman bases the separation on the supposed formation of contact metasomatic

deposits from "emanations" and of hydrothermal ones from liquids. This distinction seems unsound because, if the depth of formation of the average contact metasomatic deposit is 8,000 feet or more, the confining pressure on any water-gas separated from the magma involved would be 700 atm. or more, and the density of this water would be, at 500° C., 0.41 of that of liquid water at 4° C. and about 0.6 of that of water under such a pressure at its critical temperature (Kennedy, 1950, pp. 558–560). A water-gas of this density would be similar enough in its dielectric constant to act as a solvent in the same manner as liquid water, the lesser density being largely compensated for by the greater chemical activity inherent in higher temperatures. This relationship between density and solubility is shown graphically by Gillingham (1948, p. 269), whose figure 6 indicates that the slope of the solubility isobars at and above 700 atm. and above the critical temperature is positive. The slope of the isobars is less positive between 700 and 1,000 atm. and above the critical temperature (375° C.) than below that temperature but, nevertheless, is positive. Above 1,000 atm. the slope of the isobars apparently is the same above and below the critical temperature. It follows from this and the inability of tenuous gases to dissolve appreciable amounts of nonvolatile substances, even at high temperatures, that the tremendous volumes of contact metasomatic replacements must have been accomplished by solutions (whether gases or liquids) dense enough to carry away the debris of replacement and essentially indistinguishable in the results of their chemical reactions from hydrothermal liquids.

It would be, of course, perfectly sound classification procedure to separate deposits formed by hydrothermal fluids under high pressure and above their critical temperatures from those formed below, were criteria known for so doing. Since such criteria are unknown at present, contact metasomatic deposits should be included under those formed by hydrothermal processes, and Bateman's classification should be modified even further than he has done in the second edition. This point was made nearly twenty years ago by Loughlin and Behre (1933, pp. 47–49); and, following them, it would seem reasonable to distinguish not between contact metasomatic and hydrothermal deposits but, rather, between high-intensity hydrothermal (hypothermal) deposits formed in calcareous rocks, on the one hand, and in noncalcareous rocks, on the other.

In chapter v, section 1, in which Bateman discusses the process of "magmatic concentration," little has been changed from the first edition. The author says that the magmatic ore deposits "are magmatic products that crystallize from magmas," being "either the whole igneous mass, or part of it, or form offset bodies." Such a definition should place pegmatites (or at least those of economic value) in the "magmatic concentration" subdivision of the author's classification; and he does put them there, although some probably should be included under his II A ("Residual Liquid Segregations") as well as under his II B ("Residual Liquid Injections"). The discussion of pegmatite formation is in chapter iv, however, and contains no mention of the work of Smith (although his 1948 paper is cited at the end of the chapter), and no reference at all is made to the monograph of Cameron, Jahns, McNair, and Page (1949), which makes so important a contribution to the understanding of pegmatites, until the reading list on mica on page 758.

Bateman's definition of magmatic concentration would seem to exclude all ore bodies formed by hydrothermal processes, no matter how high the temperature and pressure of the ore-forming fluids. It is therefore logical to see such deposits as the titanium-low magnetites at Lyon Mountain, New York, and Dover, New Jersey, so classified on page 524, as they almost certainly are hydrothermal replacements and not magmatic concentrations (ore pegmatites) as the titanium-rich magnetites of Sanford Lake seem to be.

In short, Bateman, though he does not specifically say so, apparently considers that magmatic deposits come from water-poor fluids and hydrothermal from water-rich. It is to be regretted that he says nothing of the reasons for the separation of such fluids from one another in the late magmatic stage advanced by Smith (1948). Just as liquid immiscibility is the cause of the separation of sulfide melts from silicate melts in the early stages of their crystallization cycle, so it may well be the reason for the late magmatic stage separation of water-rich and silicate-rich fluids.

Much of the remainder of chapter v has been greatly improved by rearrangement and rewriting, although, after Blanchard's (1944) letter, it is surprising that there have been no qualifications made in the discussion of gossans and cappings. Chapter v, section 9, retains the rather unfortunate grouping of truly meta-

morphic mineral deposits with mineral deposits of hydrothermal origin found in metamorphic rocks.

The remaining chapters of Part I have not been changed greatly, and many typographical errors have been corrected. One or two new misleading or incorrect statements have crept in, such as the statement in chapter iii (p. 28) that solution is generally exothermic and precipitation endothermic, when the reverse is true. On page 36 the statement is made that digenite, upon cooling below 78° C., unmixes to chalcocite and covellite. Actually, Buerger's (1941) diagram shows that digenite formed above 78° C. varies considerably in composition from stoichiometric proportions and below that temperature will exsolve any excess chalcocite or covellite but not both. Digenite is not, as Bateman's statement would indicate, unstable below 78° C. On page 38 the impression is given that digenite formed above 78° C. may contain dissolved covellite but, by inference, not chalcocite. Buerger demonstrates that chalcocite may be dissolved in digenite as well.

In chapter iv, no mention is made of the work of such men as Goldschmidt, Wickman, and Osborn. Their papers go a long way to explain not only why the order of crystallization in igneous magmas is what it is but also why the constituents of the ore-forming fluids and pegmatitic fusions remain in the late-stage fluids. A sound hypothesis to explain the late separation of these two fluids from each other could have been put forward, based on the work of these men and supplemented by that of Graton and Smith. It also seems to me that this chapter contains too great an insistence on the gaseous and probably acid character of the ore-forming fluid on such occasions as it may be above its critical temperature. Kennedy's work (1950), already mentioned, shows that the density of water-gas under 700 atm. or more of confining pressure is of the order of magnitude of that of water, under the same pressures, at its critical temperatures. Such a gas would be capable of dissolving a very considerable amount of nonvolatile material, and its content of strong cations and weak anions and its ability to accomplish replacement reactions undoubtedly would be much higher than that of the tenuous gas envisioned by Fenner.

In chapter viii, the subject of geochemical prospecting is dismissed with four lines on page 393 and a reference on page 397 to the recent progress report of Hawkes. Of course, Hawkes's work contains a complete bibliography, but the importance of the work he summarizes surely merits a somewhat longer discussion in Bateman's book. In the second edition, Part II is again devoted to short descriptions of the principal metallic mineral deposits of the world, and each district described is still grouped under the metallic element which is produced from it in the greatest abundance or from which it derives the bulk of its income. The changes in Part II are much less extensive than those in Part I. Only a few deposits not considered in the first edition have been added, and all but one or two of these are dismissed with a sentence or two. The lists of references have been expanded and brought up to date, and the descriptions of a few of the districts, notably Sudbury, have been modified. By and large, however, the changes are hardly notable enough to have justified the expense of resetting Part II unless the original plates were no longer in existence. The lack of footnotes or reference numbers keyed to the reference lists, commented on by Bastin in his review of the first edition, is most unfortunate throughout the book and is particularly so in Part II. It must be admitted that the use of footnotes would add to the size and cost of the book, but they would also add markedly to its usefulness. To some extent the inclusion in each list of the appropriate general references, such as *Copper Resources of the World* and *Structural Geology of Canadian Ore Deposits*, enables the student to find a partial bibliography on nearly any deposit in which he may be interested. Some of the general references, however, are quite old, and anyone consulting, for example, Emmons' *Gold Deposits of the World* for data on Cripple Creek would be in ignorance of Koschmann's 1941 publication, unless he made a rather careful search of the bibliographies. Since the author, presumably, has already located the paper—although he does not present Koschmann's 1941 views in his discussion of the Cripple Creek ore body—it would seem reasonable to record it and save the student the duplicate effort. Another inconvenience to the reader is the mention of some author by name in the body of the text without identifying him there or in the reference list. As an example of this, on page 447 appears the statement: "Fisher shows that the carbon . . . was introduced by replacement," without any clue as to which Fisher (1938–1939) is meant or where or what he published.

The descriptions of the various deposits are almost uniformly good, although most of them are too short to give the reader more than the barest

outlines of their geology. It has been my experience that students learn more from a rather detailed study of a few typical deposits than from a brief consideration of a large number, no one of which they really understand. This is not a characteristic peculiar to Bateman's book alone; all the major texts on economic geology are similar in this respect.

With the introduction of "hydrothermal processes" into Part I, it is surprising that the compilations of "kinds of deposits and origin" preceding the descriptions of the deposits of the various metals do not mention hydrothermal deposits but only replacement and cavity-filling ones. This is not a vitally important point by any means, but it does show a lack of consistency. Another strange thing is the failure to include the name "sphalerite" in the list of zinc minerals.

One discussion in which the emphasis has been markedly changed is that of Sudbury, which ends by saying that "the staff geologists consider that the deposits were formed by hydrothermal solutions by . . . replacement, and the evidence certainly supports this view." My own discussions with the present staff geologists and my examinations of the deposits under their guidance lead me to believe that most of them think the sulfides were introduced in the molten state (as Bateman himself suggested in 1917) and that the evidence supports this view. Even at Falconbridge, where the results of hydrothermal alteration are impressive, the ore is later than the alteration. It is true that this opinion has not found recent expression in the literature, but, until it does, its acceptance at Sudbury should be mentioned.

Part III retains the same organization as in the first edition of the book, one based, except for coal and petroleum, on the use of the materials discussed. If the organization were similar to that of Part II, the primary divisions would be, instead of the metals, the nonmetallic minerals and mineral fuels of economic importance, and the emphasis in the descriptions would be on geologic occurrence in each instance instead of on use. This would seem to be a more consistent approach and one which would probably be of more benefit to students of economic geology.

The total number of figures in the new edition is increased by one over that in the old. A dozen of the figures are new or have been redrawn, the rest remain the same. The usability of the book is greatly increased by indicating the principal reference to the more important subjects in boldface type and by putting an abbreviated designation of the appropriate chapter in the upper right-hand corner of each left-hand page. The superb paper, composition, and reproduction of the cuts is a credit to the requirements of the author and the ability of the publishers. The price of the book is, for these times, moderate.

I cannot end this review without emphasizing that the comments that I have made about *Economic Mineral Deposits* are concerned with only a small fraction of the book, the dominant bulk of which is uniformly excellent and reflects only the greatest credit on the author. Economic geologists, present and future, owe him a great deal for the tremendous amount of time and effort he has devoted to what can only have been a labor of love. Finally, I should like to refer the reader to the fine review of *Economic Mineral Deposits* by E. N. Cameron (1950), which in the main considers the book from a different approach than mine and also which treats many points which I have omitted for want of space or as beyond my competence to discuss.

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Henry Gross and His Dowsing Rod. By KENNETH ROBERTS. Garden City, N.Y.: Doubleday & Co., Inc., 1951. Pp. 310; figs. 6; maps 2. $3.00.

With apologies to the ghost of Will Shakespeare, there are more things in heaven and earth, Geologist, than are dreamed of in your philosophy. Likewise to that of Sir Phil Sidney, here is a tale that will hold young geologists from play and old ones from the chimney corner. Although its well-documented statements cannot be challenged without calling scores of named, reputable people prevaricators or credulous dupes of clever hoaxing, the feats of Henry Gross with a dowsing rod remain almost incredible. Dozens of scoffers, also named, appear during the succession of amazing episodes, many with ingeniously contrived tests to trap the gifted dowser. Some scoffers "remained to pray."

During four years (1947–1950) of association with Kenneth Roberts, Henry Gross, fiftyish, discovered that he possessed divining powers far exceeding his previous knowledge, which began at age twelve, that he could dowse accurately for water underground. His learning steps progressed rapidly from that simple finding of water in the ground directly beneath him. He first learned that depth to the water can be found by walking rapidly backward away from the "vein" at a right angle to its course. The rod will dip again when the horizontal distance so traversed equals the depth.

He then learned that, if the tip of the rod were touched to "anything at all," it would then "work" on that particular substance. Thus when "touched to rye whiskey, it would work only on the rye, not on bourbon, scotch, blended whiskey or any other liquor." In succession, it would identify any one of the others. He soon discovered that there was no need to open a bottle—at least for the purpose of this experiment. The rod worked accurately when the tip was touched to sealed bottles.

Next he found that he could determine the depth, direction of flow, and discharge volume of a "vein" by *asking* the rod! " 'Is there a quart a minute flowing at the present time?' The rod dipped, meaning Yes. He poised it again and asked 'Are there two quarts?' The rod dipped. It dipped again at three quarts but refused to dip at four quarts. So Henry tried 3½ and received an affirmative answer; 3¾ and got No."

He then tried asking the rod when poised above a "vein," "Is this water good to drink?" If "excellent" the rod moved down "violently," but only "firmly" if it was "good," "hesitantly" if not particularly good, and of course, would not dip at all if the water was polluted. "Only Henry's rod can tell the quality of underground water. No engineer or geologist can do it."

Continuing his research, Henry soon successfully located "veins" in places where he had never been, merely by pointing his rod at a *photograph* or a *map*, and asking questions!

But he also learned that, for other substances than water in the earth and for the pendulum method of dowsing, especially in ascertaining the sex of an unborn child, error frequently attended his findings. Every dowser is warned to study his own performances carefully. If no water is found where the rod indicates, "it's merely that the dowser hasn't sufficiently investigated the substances responsible for the rod's movement." He may not be gifted for water-finding but may be infallible for some ores or for oil! "Henry's rod will answer any question he chooses to ask it—Is there underground water on the planet Mars? Will it rain next Saturday? Will there be war with Russia within the year?" That rod is really on a spot for *it cannot refuse to answer nor*, unless asked "Do you know—?" *can it admit ignorance.* We have no record of its ever being so asked.

Yet Henry had a few successes with nonaqueous subjects. He found an outboard motor which had been dropped in deep water a year before. To a girl scout's "pop-eyed amazement," his rod led him straight to her lost fountain pen. He found four people who deliberately challenged him and hid themselves on Roberts' Maine farm; indeed, he found a fifth, a trespasser! He has learned, however, that he should limit his queries to water in the ground, flowing under "natural conditions."

Henry's climax, to date of publication, has been the location of four "domes" of fresh water on perennially thirsty Bermuda by dowsing a map of the island when at home in Maine, 800 miles distant. Roberts has a residence on Bermuda, but Henry had never been there. Following this, he went to Bermuda and pinpointed the "domes" in the field, learned the depths to water more accurately than the rod had ad-

vised him in Maine, the depths from which the water rose, the number of radiating "veins" from each "dome," and the yield from each "dome" top. He was warned by the rod that one "dome" was polluted. The rod then found the outhouse responsible!

Drilling followed over three "domes" of good water, which was found at the predicted depths, although its top was 3 feet below sea-level in one case and 23 feet below in another. The diameter of one "dome" at the top was 6 feet. The ignorant driller's hole, slightly off-center, yielded water a trifle salty. Yields could not be accurately determined because of lack of adequate pumps. A maximum of 44 gallons per minute was measured from one well. The water was free from *B. coli*, soft to medium hard, and in two cases very low in chlorides. This drilling occurred in early 1950 and is to be credited to K. Roberts' initiative > Bermuda inertia.

Geologists may grin a bit wryly at Roberts' acid disapproval of them and the "Brotherhood of Closed Minds," but they will not readily forgive his serene and abysmal ignorance of the most elementary principles of ground-water hydrology and of the very existence of geomorphic histories and of structural and stratigraphic complexities. His and Henry's "domes or pipes" are fantastic concepts born of poor, but presumably honest, parents: ignorance (of geology) and faith (in the rod's omniscience and truthfulness). Water rises in "domes" because of "the same sort of pressure that drives up oil or that in times past has driven up mineral deposits or rocks in a state of flow." One of Henry's three Mount Washington "domes" rises from a depth of 57,500 feet and feeds 46 "veins." It terminates 126 horizontal feet from the Summit House at a depth of 50 feet and would yield, if tapped here, 128 gallons a minute. Another Mount Washington "dome," rising from 56,000 feet, possesses 51 radiating "veins," the topmost being only 26 feet below the ground surface. Total discharge of the 51 is 156 gallons a minute of "flowing" water. This quarter-million of gallons daily, piped by nature to the mountain top, is still ignored by the Summit Hotel management, which continues to bring its water up from the old source, 3,100 feet lower down the mountain slope.

The four Bermuda "domes" bring up fresh water from depths as great as 7,100 feet, halfway down to the surrounding ocean bottom. Two more Bermuda "domes" have since been found by dowsing, both terminating offshore beneath the coastal water but sending some of their radiating "veins" inland!

The Ghyben-Herzberg principle apparently does not explain this fresh water under cavernous Bermuda. Dr. Heinz Lowenstam suggests to the reviewer that the much greater Bermuda of eustatically lowered oceans during Pleistocene glaciations may have collected a body of ground water, parts of which became sealed off and still remain. But many earlier drillings have been made on the island without finding fresh water. Accepting the Bermuda account as correct, how, Geologist, did Henry locate his well sites?

Despite the enormous vertical lift involved, "domes or pipes" leak off into "veins" only at shallow depths beneath the present land surface. "Veins" may then leak off into adjacent ground and produce "seepage water," which fools some dowsers, but not Henry. "The depth of the veins has nothing whatever to do with the water table of the locality." The fact that volume of "vein" discharge into a well may vary with the season ("winter rains had forced the full strength of the vein back into its original course," etc.) is stated repeatedly but has no significance for Gross and Roberts.

Although "veins" consistently diverge or radiate from "dome" tops, Henry has found in most of his dowsings that these "veins" then converge and unite down-current (downhill). In spite of (1) this radial divergence of "veins" with increasing distance from a "dome" top and (2) this reduction in their number through coalescence, the valley-bottom farms examined by Henry are well supplied with numerous "veins." Roberts' farm has a total of 22 under less than 10 acres. They are all fed from four "domes" spaced 150–300 feet apart and rising from a maximum depth of 3,500 feet. Their "veins" radiate at depths of 16 feet or less. Six of them then converge and unite at a single point. "No geologist . . . could have mapped the strangely erratic courses" shown on Roberts' map. Henry can trace "veins" unerringly across country, but his research has not yet given us a scaled, three-dimensional depiction of these puzzling patterns and proportions.

"Veins" emerging at the surface constitute springs which Roberts emphatically insists are not ground water. "Water tables are supplied and maintained by domes of water on hilltops and mountaintops, no water dome depends in any way on any water table anywhere." But the founders of the "dome" theory attach no genetic significance to this hilltop habitat.

Neglecting quotations and two chapters not written by Roberts, the word "vein" appears at least 452 times in the 310-page book, "dome" 101 times, "bedding" or "layer" or "ledge" or "strata" 24 times, "limestone" 14 times, "seepage" 12 times, "water table" 9 times, and "ground water" twice. "Porosity" (meaning permeability) appears 4 times; so do "sand" and "gravel." "Sandstone" is used 5 times, "clay" 9 times, "quartzite" and "sand" twice each. Appearing but once are the words "fault," "structure," "oil pool," "cavern," "sink," and "granite." "Hydraulic [meaning *hydrostatic*] pressure" appears once; so does "Pleistocene" and "kaolin," but all three words are in quotations from letters of a correspondent. This completes the list of geologic terms used. What if he did write "septic" for "aseptic" every time, and Doubleday's editors let it pass? The context puts the reader right.

Would you be interested in seeing the names of some discredited scientists? Discredited for published statements are A. J. Ellis, *The Divining Rod* (U.S. Geol. Survey Water Supply Paper 416); J. W. Gregory, *Water Divining;* O. E. Meinzer, chief, Ground Water Division, U.S. Geological Survey; R. A. Millikan, president, California Institute of Technology, Nobel Prize winner in physics; A. E. Verrill, *The Bermuda Islands*. Discredited for a conversational remark is Nobel Prize winner A. A. Michelson. Discredited for a newspaper controversy with the author (in which neither would accept the conditions of the other's challenge to a field test) is Professor Ray Koon, department of horticulture, University of Massachusetts. Discredited for doubt or disbelief after a field conference and a demonstration by Henry are Dr. Gardner Murphy, head of the American Society for Psychological Research, and Professor Joseph Trefethen, state geologist of Maine.

A conditional passing grade is given, by courtesy, to Dr. Vannevar Bush, president, Carnegie Institution of Washington; Dr. Bradley Dewey, colloid chemist; and Professor J. B. Rhine, department of parapsychology, Duke University.

Three scientific men are given full approval: Professor Charles Richet, University of Paris, Nobel Prize winner in medicine and physiology; Dr. Arthur Brown, deputy director of a Navy Research group; and Dr. Horace Levinson, chairman of a National Research Council committee. Only ignorance of recent dowsing literature can account for the absence in this list of the names of Professor S. W. Tromp, Fouad I University, Cairo, and his sympathetic reviewer, Professor Alan Bateman, department of geological sciences, Yale University.

Professor Marland Billings, department of geology, Harvard University, is mentioned in the Bermuda account but fortunately escapes both condemnation and commendation.

Dr. Albert Einstein narrowly misses inclusion somewhere in these categories, for Henry has gratuitously dowsed the Einstein residential property in Princeton and found two "veins" only 8 feet below the surface. Apparently no further investigations have been made and luckily excavation for the basement of the house missed them.

The rest of us are told, as a group, "that dowsers know things that geologists can neither know nor explain; and that geologists will fight to the death to refute their lack of perception." "At last the shoe is on the proper foot; the geologists have been the misrepresenters; the dowsers have been right." Investigation of whether or why dowsing rods work is time and effort wasted. "Rods [do] work and nobody can find out why." Alas, that so many New England farmers, "suffering from empty wells and parched fields," have believed "the half-truths so freely spread abroad by geologists."

Probably the outstanding challenge of this book to a geologist's curiosity is the source that supplies the rising "spouts," "pipes," or "domes" of subterranean water. Roberts gives us no hint, apparently does not have that curiosity and has not asked Henry to ask the rod. Bermuda, 600 miles from the North American shore, has six such "domes." Mount Washington's summit area, a mile above tide, has three, and they rise from maximum depths of nearly 10 miles below sea-level. This unmineralized water has the grace to start at a temperature of 141° (presumably Fahrenheit). It cools to 33° somewhere along the 11-mile upward journey. "I feel profoundly sorry for geologists," says Roberts. "They are missing something enormous."

Enthusiastic (etymologically, "God-infused") author Roberts is confident that unconscious muscular movements cannot be the explanation of Henry's performances, nor can extra-sensory perception (although Rhine tested Henry and went away convinced that ESP was the explanation). "Emanations," however vaguely defined, do not cause the reaction of the Gross rod. Henry and other accurate dowsers are

agents possessing and demonstrating a "Force" which scientists are bullheadedly ignoring or denying, a "Force" which, "properly utilized, would prevent wars, move mountains, turn deserts into lands of plenty, feed the hungry, cure the sick and change the face of the world."

Bermuda may be the present climax of this Gross and Roberts research, but the future is bright with promise. Henry, now incorporated as "Water Unlimited," has already long-distance dowsed New Providence Island in the Bahamas, Capri in the Mediterranean, and the Sahara Desert. He has found "domes" in these dry lands, both beneath the land and beneath the adjacent sea. The latter have landward-flowing "veins"! The Atlas Mountains "dome" outdomes all "domes" ever found to date. Although the Mediterranean averages less than 200 miles distant to the north, a supercolossal "vein" (really "something BIG," said Henry; and "a river rather than a vein," wrote Kenneth) flows south from these mountains, completely across under the desert to discharge as a submarine spring in the Gulf of Guinea 1,000 miles away. These amazing discharges of deep subterranean fresh water, rising thousands, even tens of thousands, of feet against gravity beneath both land and sea, then inexplicably succumbing in a burst of radiating veins when almost free from the lithosphere, make clear to the reviewer why the ocean is brimful and overflowing but leave unsolved the mystery of its saltiness.

In printing this book, Doubleday and Company appears to be knocking for admission to that select circle of uncritical publishers which Macmillan recently joined by bringing out Velikowsky's *Worlds in Collision*.

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Stratigraphy and Sedimentation. By W. C. KRUMBEIN and L. L. SLOSS. San Francisco: W. H. Freeman & Co., 1951. Pp. ix+497; figs. 122. $5.00.

The principles and procedures that relate sediment deposition to the physical history of the earth are the subject of this well-written text.

The discussion of classification of strata is based on recent experiences and follows the recommendations of the Stratigraphic Commission. Field and laboratory techniques are emphasized; treatment is more comprehensive than generally available except in books on field methods. The summary of subsurface methods will be particularly enlightening to many students.

The succeeding chapters on rocks, processes, and environments are adequate summaries of matter available in less abridged form in journals and other texts. Sedimentology has acquired several vernacular terms and has assigned varying definitions to them. The terms "clastic" and "detrital" are presented as synonymous with "terrigenous" and "noncalcareous," but there is contradiction in referring to "clastic limestones"; many "clastic" particles have not been formed by the breaking of rocks, whereas some of the "nonclastics" have. Thus "clastic ratio," as applied by the authors to the ratio of sediments derived from erosion of preexistent rocks to the total of limestones, dolomites, and evaporites, is somewhat misleading. Inquiry has shown that geologists use the term "detritus" for any sediment that is capable of drifting or particle-rubbing, as implicit in the word, regardless of composition or origin. The term "terrigenous" has been adopted and preferred by some for sediments eroded from rocks, but this usage is not universal. The writers further recognize the inconsistency of referring to all nonmarine sediments as "continental." Loose usage of common words confuses the student, as well as all geologists; but this text is as consistent as other texts and common usage.

Stratigraphic paleontology, sedimentary facies, and the principles of correlation are topics that are little considered in texts on sedimentation, for they are fundamentally stratigraphic—and there has not been an adequate summary of stratigraphic principles for many years. The book by Krumbein and Sloss has much on these topics that is novel and thoughtful and contains many suggestive and representative maps and sections.

The succeeding chapters on the tectonic framework of sedimentation and sedimentary tectonics are the most significant contribution of the text. Though one may not agree with some statements of the authors, their basic emphasis is excellent, and the principles set forth are those that need greater consideration. The discussion of the dependence of sediment character on tectonism is the best that has appeared in a textbook. The principles help the student acquire an orderly comprehension of the significance of the stratified rocks. If the following re-

marks seem to exaggerate the shortcomings of the book, it is because they are presented in the hope that such criticisms will prove more helpful than the commendation and praise that are warranted for all but a few paragraphs.

A summary of the development and classification of geotectonic elements begins the discussion. Unfortunately, the authors do not clearly distinguish between the present structural surfaces; the paleostructural or stratigraphic surfaces formed by subsidence during the deposition of their contained rocks; and the physiographic surfaces of deposition, such as those classified most recently by Rich. For example, the Michigan Basin is not an autogeosyncline but a structure; the autogeosyncline was stratigraphic, located in the area of the present structure but encompassing only the part of the Paleozoic later than the middle Silurian, as the authors state on a subsequent page. The text repeats a common fallacy in placing a topographic or stratigraphic subsidence in the area of the present structural basin in Michigan in the Niagaran (fig. 11-6); recent studies have shown the series to be thinnest there (Cohee), in a reef belt transecting present structure (Lowenstam). Though the authors tend to use "geosyncline" in a stratigraphic sense, occasionally they give it a geomorphic meaning, as (p. 374) in referring to black shales as laid in the "deeper" portions of geosynclines—actually in deeper water, which was far from the axis. Again, Tercier's classification in which "geosynclinal sedimentation" is used in the physiographic sense is listed without comment (p. 385), though it has been properly criticized for designating classes by terms of dissimilar categories (Lombard).

The reviewer finds after reading this text that he, too, failed to anticipate ambiguity when he defined a zeugeosyncline as "yoked" to an uplift, for he failed to recall that "uplift" has been applied to relatively little subsiding regions as well as to elevated highlands. The middle Paleozoic of the Forest City Basin and the Mississippian of the Illinois Basin are not deposits in zeugeosynclines but are in autogeosynclinal areas. On the other hand, the Newark was laid down not in zeugeosynclines (p. 337) but in taphrogeosynclines; by suppressing taphrogeosynclines to emphasize the similarities with zeugeosynclines, one masks their differences. The late Paleozoic in Colorado and New Mexico was laid in typical zeugeosynclines.

The authors quite properly emphasized the multiple factors responsible for the sediment; the presence and nature of the source land is quite as important as the behavior of the depositional site. Thus virtually all kinds of sediments are thickest in geosynclines. Great thickness implies great subsidence. An unwary student will take the observation that certain kinds of sediments are most abundant in certain tectonic settings to mean that they characterize, or define, such a setting; the word "typical" occasionally can be misconstrued similarly. However, anyone who studies the table of "tectonoenvironmental classification" (p. 388) and the associated graphic representation (fig. 12-5) must appreciate that the relations are complex.

The concluding chapters on stratigraphic maps and paleography seem refreshingly simple and descriptive. One error of terminology has been repeated from its source; "isoliths" are lines of equal rock character (Grossman), not of thickness of lithology (p. 404). The application of the principles set forth in this book to the analysis of a time and region is an excellent means of summarizing the message of the authors. The text concludes with an extended list of references to the literature and to articles that treat subjects more fully, and with a rather detailed index of subjects and authors.

The value of the book is enhanced by remarkably clear and varied typography and aptly selected and well-drafted illustrations. The binding and paper are excellent; the pages can readily be laid open at any chosen point. A book hardly could be more enlightening and challenging to those who are to study or engage in stratigraphic geology.

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Dating the Past: An Introduction to Geochronology. By FREDERICK E. ZEUNER. 2d ed. New York: Longmans, Green & Co., Inc.; Frome and London: Butler & Tanner, Ltd., 1951. Pp. xviii+474; figs. 103; pls. 29. $5.50.

The first edition of this book appeared in 1946 and was reviewed in detail by Ernst Antevs (*Journal of Geology*, vol. 55, pp. 527–530, 1947). In the new edition the text has been enlarged by 30 pages and brought up to date to December, 1949. About 100 new references appear in the Bibliography, and numerous additions and replacements have been made in charts, tables, figures, and plates. Recent information is included on such subjects as deep-sea core inter-

pretation, radioactive carbon dating, pollen analysis, and varve chronology. The absolute astronomical chronology of the glacials and interglacials of the Pleistocene again is supported in the text, and fourteen objections to it, which have been raised by various writers, are considered briefly in the Appendix.

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Soils: Their Origin, Constitution, and Classification: An Introduction to Pedology. By GILBERT WOODING ROBINSON. 3d ed. London: Thomas Murby & Co.; New York: John Wiley & Sons, Inc., 1949. Pp. xxi+573; figs. 22; pls. 9. $5.00.

The first and second editions of this book by Professor Robinson, of the University College of North Wales, appeared in 1932 and 1935, respectively. Because of its well-balanced treatment and its emphasis on the natural-science aspect of soils, it is particularly useful to scientists in allied fields. As expressed by the author, the book was written primarily for "those who are interested in the soil as an object of study in itself, and secondarily for those whose interest lies in its economic or geographical significance."

The new edition has been enlarged from 442 to 573 pages, and 5 new figures and 4 new plates have been added. More important, the text has been consistently revised and brought up to date. Much of the new material is included in the chapters on "Soil Formation," "Soil Classification," and "Soil Surveys." There is little change in the over-all organization. The first part of the book deals with the physical properties of soils and factors in soil formation. This is followed by a discussion of the major soil groups and, in concluding chapters, by a consideration of soil classification, geography of soils, soil surveys, soil analysis, and soil use.

As a geologist, the reviewer is impressed by the circumstance that a large part of the knowledge of soils is based on youthful soils developed on late Pleistocene parent-materials. Much less information is available on mature profiles, which probably were normal to the geologic past and should be used as a basis in interpreting ancient sediments.

LELAND HORBERG

University of Chicago

Soils of the Netherlands. By C. H. EDELMAN. Amsterdam: North Holland Publishing Co., 1950. Pp. vii+177; figs. 67; provisional soil map. Fr. 17.50.

Prepared for the convenience of the members of the Fourth International Congress of Soil Science, held in the Netherlands and Belgium during the summer of 1950, *Soils of the Netherlands* summarizes soil conditions in the Netherlands and emphasizes the ways in which soils have been modified by man. A provisional soil map, in fifty-five colors, and sixty-seven line drawings and photographs illustrate the text. Geologists will appreciate the discussion on relationships of Pleistocene and "Holocene" deposits to soil formation and the emphasis on the time factor in soil formation, even though these topics are only incidental to the main objective of the book.

The first chapter presents a concise general description of the Netherlands and a geological time scale to cover the period since the retreat of the last glacier from northern Europe. The soil classification system used in the Netherlands is compared briefly with that used in the United States.

Soils are discussed under twelve main heads, based on the mode and locale of deposition of soil materials. Edelman explains how the sands of the Netherlands have accumulated partly through sedimentation by glacial streams and partly through the wind action on these deposits. The discussion of the "river clay soils" is as much a lesson in sedimentation as it is one in pedology, but the author shows how soil-forming processes begin to work on the sediments and how profile development advances.

Much attention is paid to the peat deposits and their place in Pleistocene and Holocene geology. Many of the peat beds have been excavated for fuel, and the underlying mineral sediments converted to productive soils. Where sediments beneath the peat are very sandy, it is impossible to reclaim them for farming, because water seeps in more rapidly than it can be pumped out; but clayey sediments, exposed by removal of peat, permit little seepage and can be reclaimed for agriculture. In early stages of reclamation, some of the noncalcareous sediments become exceedingly acid through oxidation of ferrous sulfide to sulfuric acid. Reclamation of soils of this sort is slow, expensive, and difficult.

Edelman describes how old sand dunes, behind the young dunes of the sea coast, are excavated, leveled, ditched, and tiled, so that the water table can be maintained at exactly 55 cm. The old dunes have almost no value, but the new land, with its water table controlled at 55 cm., is among the most valuable for farming of any land in the world. The surplus sand is used for town foundations and for construction.

Dutch scientists have obtained more detailed information on Pleistocene and Holocene deposits, perhaps, than is available in any other country because, as Edelman points out, such knowledge is absolutely essential to the sound development of Netherlands agriculture and reclamation of land and sea bottom for farming.

Edelman's book gives basic data needed for estimating the time requirement for the development of certain kinds of soils. For example, the strongly developed Heath podzol profiles are formed in "cover sands," many of which are estimated to have been in place less than seven thousand years. Acid, light-gray podzol soils have been changed to black fertile soils in a few hundred years through intensive manuring, liming, and use of commercial fertilizers. Many kinds of "artificial" soils have been formed through mechanical mixing of various kinds of peat with different types of mineral sediments.

This book will impress the reader with the importance of man as an agent in sedimentation and soil formation in this small but important corner of Europe. There is a detailed Table of Contents and short bibliographies for several of the chapters, but no index.

JAMES THORP

U.S. Department of Agriculture
Division of Soil Survey

Die Erzmineralien und ihre Verwachsungen. By PAUL RAMDOHR. Berlin: Akademie-Verlag, 1950. Pp. xvi+826; figs. 431. DM. 88.

Lehrbuch der Erzmikroskopie, by Schneiderhohn and Ramdohr, the second volume of which appeared in 1931, is out of print, and the last copies were destroyed in the air war. Professor Schneiderhohn is pressingly absorbed in other duties, and there has fallen to Dr Ramdohr the task of revising and largely rewriting this book, which has been of great value to students of ore deposits the world over. The title, however, has been changed, as the present work does not include a consideration of microscopic methods, which constituted the first volume of *Lehrbuch der Erzmikroskopie*.

The revised work was accomplished under the great difficulties of war and reconstruction times. The first part, dealing with the c asses of ore deposits, paragenesis of ore minerals, and the textural and structural features of ores, is entirely new. The second part, dealing systematically with the characteristics of the ore minerals, follows closely the plan of the earlier Schneiderhohn and Ramdohr volume but brings the material up to date. Many of the illustrations are new. Unusual textures are stressed and illustrated rather than those already familiar to most students of ore deposits.

The first 55 pages are devoted to the more general features of ore deposits, which are grouped under the four classes of meteorites, magmatic deposits, sedimentary deposits, and metamorphic deposits. The major divisions are the familiar ones of rock classification, and their use has the advantage of simplicity and of stressing the fact that ores are simply rocks of somewhat unusual composition. Meteorites are only briefly touched upon to indicate that extraterrestrial sources have contributed, however meagerly, to the earth's supply of metals. Major zoning of the earth into an iron core, an intermediate zone rich in oxides and sulfides, and an outer silicate zone shows that the processes of metal concentration go back to the cosmic stages, although only the outer silicate-rich zone seems to be directly concerned in ore deposition. Dynamo-metamorphic products are deservedly discussed at some length because they have received little systematic treatment from most students of ores. Although not a primary cause of ore deposition, dynamo-metamorphism has, in many cases, produced profound changes in texture and mineral composition. These are discussed under the headings of (1) metamorphism through temperature changes and uniform pressure in all directions and (2) metamorphism under differential pressure with or without the cooperation of temperature changes. Some of these changes have economic implications, as, for example, the metamorphic generation of magnetite from nonmagnetic iron compounds which renders an ore susceptible to magmatic concentration. All modifications of ores caused by declining temperatures following crystallization, such as unmixing and reaction rims, are classed as temperature metamorphic processes. A number of instructive illustrations are given

of recrystallization, flowage, and brecciation in ores under the influence of differential pressures.

The comprehensive class of magmatic processes is subdivided first into plutonic and volcanic, and the plutonic is further divided into (*a*) "Intramagmatic Stage," (*b*) "Pegmatitic-pneumatolytic Stage," and (*c*) "Plutonic-hydrothermal Stage." In comparing the last two stages, the author notes not only that the plutonic-hydrothermal deposits were precipitated from solutions below rather than above the critical temperature of water (374° C.) but that the transporting power of the solutions declined as they became both cooler and more alkaline, rather than acid, in quality. Notable primary changes in ore deposition with depth may result from these processes. There is no lower temperature limit for hydrothermal deposition but rather a gradation into deposits in which vadose waters play a role.

Pages 22–26 are devoted to a paragenetic table of 92 minerals of economic importance in which they are classified under 33 genetic groups. The relative importance of the several groups is indicated by different symbols.

One of the most valuable parts of the book (pp. 55–172) deals with ore textures and their interpretation. This section is profusely illustrated, mainly by halftones but in part by drawings. The internal characters of individual grains, such as zoning and others, in many cases made visible only by etching, are first considered. On page 63 there is a table of 50 minerals in which zoning has been noted. The table also indicates the cause of the zoning and its form. The forms of mineral aggregates, that is, the texture or fabric, are next considered. Myrmekitic or graphic textures are shown by excellent halftones. It is known that such textures may be formed in at least nine ways and possibly more. On pages 84–86 are listed, with references, 97 mineral associations that have been reported as graphic intergrowths.

Fourteen pages are devoted to replacement textures and twenty-one to unmixing textures, including a seven-page tabulation of described examples attributed to unmixing. All except four of the 120 occurrences cited in this table seem to conform to the principle which the reviewer (1950, p. 12) has emphasized, that the products of unmixing are usually, if not always, minerals possessing definite mineralogic or chemical similarities. The last four examples cited by Ramdohr are associations of olivine with magnetite or with ilmenite, of garnet with magnetite, and of bronzite with ilmenite, that is, of minerals of entirely distinct classes, silicates with oxides. It may well be that these four cases are not results of unmixing.

Pages 135–144 are devoted to colloidal textures. Certain arborescent or mosslike growths of metallic minerals in a matrix of crystalline calcite (p. 143) are interpreted by Ramdohr as indicating that the matrix was originally colloidal calcium carbonate. In the absence of any specific textural evidence that the matrix was once colloidal or, indeed, that colloidal calcium carbonate is ever produced by ore deposition, it would seem that the alternative hypothesis should be considered, namely, that metallic mineral substances in solution may diffuse into readily replaceable *crystalline* rocks, such as limestones, and assume their characteristic forms without appreciable control by directional features of the calcite, such as cleavage and grain boundaries. The growth of pyrite cubes transecting the foliation of a schist is evidence that even rocks composed of many silicate minerals may be replaced without any considerable control by textural features. Ores of the cobalt-nickel-silver type seem to furnish excellent examples of the formation of skeleton crystals and tubercle-like growths by diffuse penetration of crystalline carbonate-rich wall rocks (Bastin, 1950, pp. 42–43).

Pages 145–154 are devoted to the practical applications of ore microscopy to the problems of milling of ores.

A short section is devoted specifically to the criteria for determining the genetic position of ore deposits. Certain minerals that have been found empirically to be characteristic of certain ranges of temperature, pressure, and concentration are termed "typomorphic." Wurtzite, for example, is commonly confined to the low-temperature, metasomatic lead-zinc occurrences and is typomorphic of such deposits. Certain groups of minerals may have genetic significance even if an individual mineral has not; they are termed a "typomorphic paragenesis." Thus associations of pentlandite and pyrrhotite with chalcopyrite and with or without magnetite are typomorphic of orthomagmatic genesis. In analogous fashion we may speak of "typomorphic structures," such as certain unmixing textures and "gel" textures. A table on pages 156–158 classifies typomorphic features with respect to ore classes. It is interesting to note that Ramdohr regards the iron-rich sphalerite, in general, as a higher-temperature form than the

iron-poor varieties. Seven pages are devoted to ore minerals and paragenetic sequences as a "geologic thermometer." The uses of melting points, inversion points, unmixing, and other phenomena are discussed.

Pages 175–775, constituting most of the volume, comprise the "Systematic Part," in which the characteristics of 343 ore minerals are discussed. Gangue minerals are only briefly treated. Subheadings which may not apply to every ore mineral include (1) "General," (2) "Behavior on Polishing," (3) "Reflecting Power," (4) "Behavior on Etching," (5) "Physical-chemical," (6) "Texture," (7) "Special Textures," (8) "Diagnosis," (9) "Paragenesis," (10) "Studied Occurrences," (11) "Publications," and (12) "Powder Patterns" (X-ray). The data on behavior under etching is based mainly on the standard text of M. N. Short, supplemented in some cases by data from Murdoch and from Davy and Farnham. Ramdohr cites a case in which, out of a paragenetic sequence of 12 ore minerals, only four could be certainly identified by etch tests. Although etch tests are an appropriate starting point in the identification of ore minerals, Ramdohr believes that they should be supplemented by optical methods, microchemical tests, and X-ray powder patterns.

The volume ends with a Bibliography of 804 titles, a locality index, and a subject index.

Professor Ramdohr's book is undoubtedly the most complete and authoritative source of information available on ore minerals, their microscopic characters, and their interpretation. It will be an invaluable aid to all advanced workers in this field.

REFERENCE CITED

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COMMUNICATIONS AND ANNOUNCEMENTS

COMMISSION ON GEOCHEMICAL LOCALIZATION OF THE ELEMENTS

At the recent meeting of the International Union of Chemistry in New York and Washington, the Commission on Geochemical Localization of the Elements, organized by the Section of Inorganic Chemistry, held its first meetings. It was agreed that one of the needs of the field that could be met by the commission was to act as a central source of information as to what research in geochemistry was being undertaken and where. It is therefore requested that persons and organizations that now have under way or that expect in the near future to undertake research projects or bibliographic projects in the field of geochemistry inform the commission of these projects and their scope. Those in the Western Hemisphere should write to Dr. Michael Fleischer, U.S. Geological Survey, Washington 25, D.C.; those elsewhere should write to Professor T. F. W. Barth, Geological Museum, Oslo 45, Norway.

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Joint meetings of the Geological Society of America, Southeastern Section, and the Southeastern Mineral Symposium will be held May 1, 2, and 3 at the Hotel Roanoke, Roanoke, Virginia, under the sponsorship of the Department of Geology of the Virginia Polytechnic Institute. The program will include a panel discussion on employment opportunities in geology, presentation of papers on such subjects as ground water, coal geology, structural geology, atomic energy as a geological tool, oil and gas possibilities, etc., a field trip covering pertinent features of the Roanoke area, and an address by Dr. Walter H. Bucher.

Inquiries should be addressed to Dr. Byron N. Cooper, Box 634, Blacksburg, Virginia.

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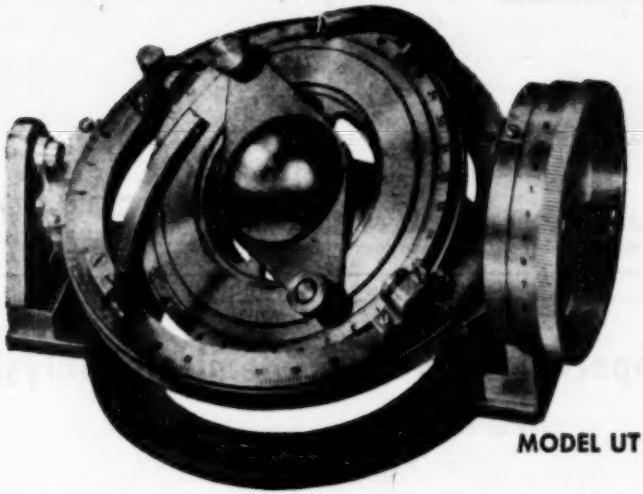

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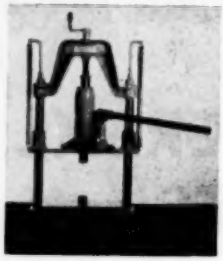

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